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THE INSTITUTION OF PETROLEUM TECHNOLOGISTS.

NOTICES.

The Institution as a body is not responsible for the statements of opinion expressed in any of its publications.

Copyright. Authors of papers and articles are requested to transfer their copyright to the Institution for a period of six months from the date of receipt of the paper. Such transfer should be made in writing when the manuscript is forwarded to the Editor. Editors are permitted to publish abstracts, providing that acknowledgment is made to The Institution of Petroleum Technologists.

Issue of Journal. The Journal is issued in six parts per volume, commencing in February of each year, with occasional extra numbers when necessary. The Title Page, Table of Contents and Index to each volume are published in the first issue of the succeeding volume.

Members whose subscription is not in arrear receive the Journal free of cost, and additional copies are charged at the rate of 7s. 6d. per part, unless otherwise stated. A member whose subscription is not paid by June 30th of the year for which it is due is considered to be in arrear.

Changes of Address. Members are requested to notify any change of address to the Secretary.

Papers and Articles. Members are invited to submit papers to be read at the General Meetings of the Institution, and are specially asked to forward articles for consideration for publication in the Journal. Diagrams, illustrations, etc., should be suitable for direct photographic reproduction.

Authors of papers published in the Journal are entitled to receive 25 free reprints of their contribution. Further copies may be obtained on payment and orders for these should be sent to the Secretary when the manuscript is forwarded to the Editor. The free reprints do not include any discussion which may be published with the paper, but authors may have such discussion included in their reprints on payment of the additional cost.

Galley proofs of the paper to be read at a General Meeting are available at the time of the meeting, but members desirous of receiving such galley proofs in advance should apply to the Secretary.

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Abstracts. Abstracts of the more important articles and patent specifications are published with each issue of the Journal, this supplement being paged independently of the transactions. Members desiring to have the Abstracts printed on one side of the paper only can be supplied with uncorrected galley proofs at a charge of 10s. per annum per copy, payable in advance.

Medals. The Redwood Medal is awarded, at the discretion of the Council, to the person who shall have made the most meritorious contribution to petroleum technology, in the form of a paper or papers published in the Journal of the Institution, during two successive sessions, preference being given to original work and to papers which have been read before the Institution and discussed. The award is not confined to members of the Institution and may be withheld if no contribution is considered to be of sufficient merit.

A medal and a prize of five guineas is awarded annually by the Council to that Student Member of the Institution who shall, in their opinion, have presented the best paper during the session.

Research Fund. The sum of £400 is allocated in each calendar year to the advancement of Research in Petroleum Technology and its basic sciences, and the Council are prepared to receive applications for assistance from this fund.

Applications from persons proposing to engage in research in any university or other teaching institution must be supported by the professor under whom the applicant will be working.

Applications from Associate Members, Students or Associates of the Institution of Petroleum Technologists and non-members, not proposing to engage in research in any university or other teaching institution, must be supported by a Member of the Institution or other responsible person.

Applications from full Members of the Institution do not require a supporter.

Applications for grants from this fund will be considered in June and December of each year and must be received by the Secretary not later than June 1st or December 1st respectively. Application forms may be obtained from the Secretary of the Institution at Aldine House, Bedford Street, London, W.C. 2.

Advertisements. Advertisements are inserted in the Journal, and information as to terms, etc., can be obtained from Mr. Thomas Tofts, 93 and 94, Chancery Lane, London, W.C. 2. (Telephone No. Holborn 8106.)

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Library. The Institution's Library may be consulted between the hours of 11 a.m. and 4 p.m. daily. (Saturdays, 11 a.m. to 1 p.m.)

ADDITIONS TO THE LIBRARY SINCE THE LAST JOURNAL.

The Geology of the Netherlands East Indies, 1925. By H. A. Brouwer.

Flame and Combustion in Gases. By Dr. W. A. Bone and Dr. T. A. Townend. 1927.

Practice of Lubrication. By T. C. Thomsen. 1926.

Low Temperature Carbonisation. By Dr. C. H. Lander and R. F. McKay.

Petroleum Tables. By Wm. Davies.

The Geology of the Salt-Dome Oil Fields. Edited by Raymond C. Moore. 1926.

Burning Liquid Fuel. By Dr. Wm. Newton Best. 1927.

Chemistry of the Oil Industries. J. E. Southcombe. 1926.

From Mr. R. D. Forrester :—

Steam Shovel Work in Tropical South America.

From the Junior Institution of Engineers :—

Proceedings. Vol. XXXVII. 1926-7.

From the American Society of Testing Materials :—

Proceedings. Vol. 27, 1927. Part. I., Committee Reports, Tentative Standards. Part II. Technical Papers.

Records of the Geological Survey of India. Vol. LX. Parts 2 and 3, 1927.

U.S.A.

From U.S. Bureau of Mines :—

Technical Paper 419 : Safe Practices at Oil Derricks. By H. C. Miller.

Serial No. 2834, Oct., 1927 : Reduction of Breathing Losses from Vapour-tight lease Tanks. By L. Schmidt.

Serial No. 2840, Nov., 1927 : The Carburetion of Combustible Gas with Butane and Propane-Butane Mixtures with particular reference to the Carburetion of Water Gas. By W. W. Odell.

Serial No. 2843, Dec., 1927 : The Sulphur Content of Commercial Motor Fuels. By A. J. Kraemer, E. C. Lane and C. S. Luce.

Serial No. 2847, Dec., 1927 : Prevention of Hydrogen Sulphide Poisoning in Handling and Refining High-sulphur Petroleums. By H. C. Fowler.

Serial No. 2850, Dec., 1927 : Stocks of Petroleum Products held by Exporters. By G. R. Hopkins.

PERSONAL NOTES OF MEMBERS AND SPECIAL NOTICES.

It is suggested that members send information regarding their movements to the Secretary, for insertion under this heading.

Mr. J. A. CHILD is now in Colombia, South America.

Mr. C. E. JOBLING is in Burma.

Mr. F. R. KELLY has left for Venezuela.

Mr. C. E. MASSEY is now in England.

Mr. J. C. TEMPLETON is now home.

Mr. W. G. LINDSAY is home from Burma.

Mr. W. J. REYNOLDS is now in Venezuela.

Mr. A. LLOYD is home from Burma.

Mr. T. G. B. DAVIES is in Burma.

Mr. W. P. JONES is in Transvaal.

Mr. F. G. FORTESCUE has now returned to Burma.

The Secretary will be glad to receive information as to the whereabouts of the following members:—J. M. BAILEY, G. M. GREAVES, D. H. GWINNER, C. E. JOBLING, K. C. JOHNSON AND J. E. NICHOLLS.

The suggestion has been made that complete bound sets of the Journal should be available for sale to members of the Institution. Any member wishing to avail himself of this opportunity should communicate with the Secretary.

The engraving of the members of the Institution, made by Messrs. H. JWhitlock and Co., is now ready, the cost being £10 10s., £7 7s. and £5 5s., together with the key. For the convenience of members, a photograph of the engraving has been made, size 14 in. by 10½ in., at a cost of £1 1s., inclusive of the key. Application for copies of the engraving or the photograph should be made to Messrs. H. J. Whitlock and Co., Cory's Building, Fenchurch Street, London, E.C. 3.

OBITUARY.

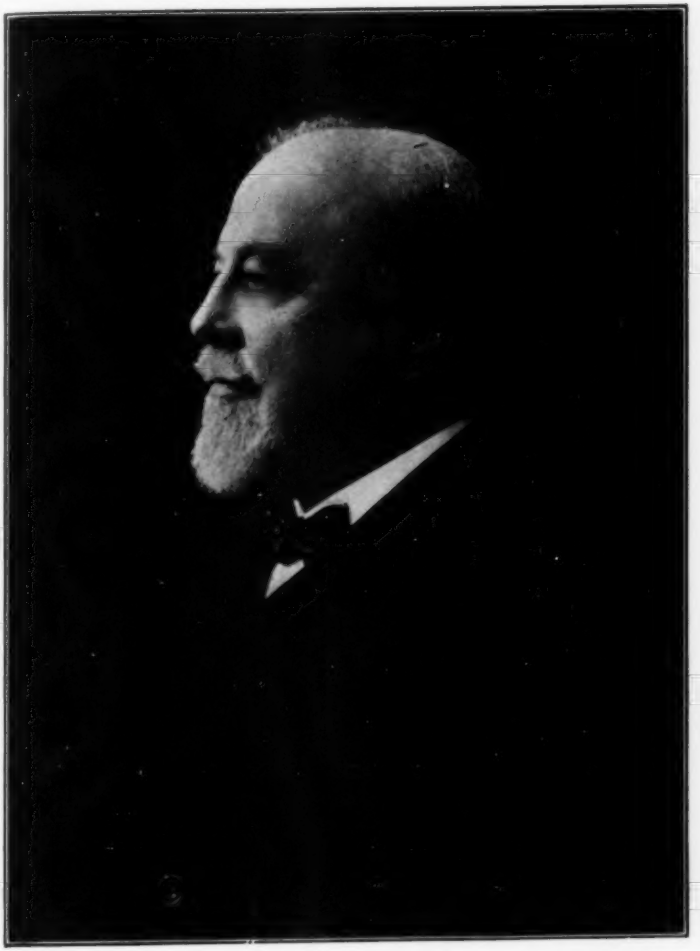
ADMIRAL SIR EDMOND JOHN WARRE SLADE, K.C.I.E., K.C.V.O., R.N.

THE death of Admiral Sir Edmond Slade on January 20, 1928, at his residence at Campden Hill, W., marks the passing of a distinguished officer and a notable figure in the petroleum industry.

Born on March 20, 1859, Edmond John Warre Slade was the eldest son of the Rev. George FitzClarence Slade, and a grandson of General Sir John Slade, Bart. Educated at Eton, he joined the Royal Navy in 1872, and became a midshipman in 1874. He was promoted to sub-lieutenant in March, 1878, and to lieutenant in December, 1879. On promotion, his first appointment was to the sloop "Fawn" in the Red Sea, where he carried out the work of a junior surveyor. In August, 1882, he joined the torpedo depot-ship "Hecla" in the Mediterranean, and served in her during the Egyptian War. In September, 1883, he was appointed to the "Vernon" for torpedo instruction, and joined the flagships of the Channel Squadron, the "Minotaur," as torpedo-lieutenant in March, 1886, transferring to the "Northumberland" when she was made flagship. He then became a staff officer in the torpedo school-ship "Defiance" at Devonport. He resumed sea service in May, 1890, as torpedo lieutenant of the "Rodney" and was appointed torpedo lieutenant of the "Camperdown," the channel flagship, eighteen months later. In January, 1893, he joined the "Trafalgar" as first and torpedo lieutenant. He was appointed Commander in 1894 in command of the paddle-gunboat "Cockatrice," stationed in the Danube on the Danube Commission, and in March, 1898, took command of the sloop "Algerine" in China. In December, 1889, he was promoted to captain, and after a term at the Naval College joined the cruiser "Diana" in April, 1902. In the following year he was created M.V.O. when King Edward visited Malta.

The course for senior officers at the Royal Naval College, Greenwich, was placed in his charge in May, 1904, and when relinquished by him had become sufficiently important for a flag officer to be appointed as his successor.

Captain Slade was appointed Director of Naval Intelligence at the Admiralty in November, 1907, being promoted to Rear-Admiral in the following year. In December, 1908, he represented the Admiralty at the International Maritime Conference, and in March, 1909, was appointed Commander-in-Chief in the East Indies. He was created K.C.I.E. for his services in January, 1911, and on the occasion of the Durbar in 1912 was also made K.C.V.O.



THE LATE ADMIRAL SIR EDMOND JOHN WARRE SLADE,
K.C.I.E., K.C.V.O.

Rear-Admiral Slade returned to England in 1912, and was appointed for special service regarding oil fuel supplies. When, two years later, the Admiralty became a large shareholder in the Anglo-Persian Oil Company, Vice-Admiral Slade, as he was then, was nominated as their representative on the board. At the time of his death he was Vice-Chairman of the board.

During the War his experience was fully utilised by the Admiralty and although he retired from the active list on his promotion to full Admiral in August, 1917, he continued on special service until the cessation of hostilities.

Admiral Slade was elected an Honorary Member of this Institution in 1915.

STANDARD METHODS OF TESTING PETROLEUM AND ITS PRODUCTS.

In view of the proposed publication during the coming year of a second edition of "Standard Methods of Testing Petroleum and Its Products," it has been decided that the remaining copies of the first edition shall be made available at 3s. per copy on and after January 1, 1928. Of this amount 1s. 6d. will be allowed as a rebate on the price of the second edition when published, and a voucher for this will be enclosed with each copy of the first edition sold after the above-mentioned date.

UNIVERSITY OF MICHIGAN.

We have received particulars of the Graduate Courses in Chemical Engineering at the University of Michigan. Facilities are available for studies and research on batch and continuous distillation, dephlegmation, gas absorption, filtration, analysis of flue and fuel gases, calorific value of fuels, and the testing of lubricants and fuels. The Petroleum Laboratory and the Fuel Research Laboratory are especially equipped for the study of motor fuels and other petroleum products, and the apparatus in the Combustion Laboratory includes facilities for studying the explosion of hydrocarbon air mixtures in closed vessels.

Full particulars of the courses can be obtained from the Registrar of the University, Ann Arbor, Michigan, U.S.A.

STANDARD METHODS OF TESTING PETROLEUM AND ITS PRODUCTS.

Report of the Standardization Committee, giving details of Methods of Test for Petroleum and Its Products.

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A brief survey of the Technology of Petroleum based upon a Course of Lectures given by Members of the Institution of Petroleum Technologists at the Petroleum Exhibition, Crystal Palace, 1920.

This work will be found of value to students and those desirous of obtaining an elementary knowledge of the Technology of Petroleum.

A few remaining copies for disposal at **2s. 6d.**

REPORT OF THE EMPIRE MOTOR FUELS COMMITTEE.

The complete Report of the Empire Motor Fuels Committee, containing valuable data relating to Fuels for Internal Combustion Engines.

A few copies are still available to members only at **1s.**

All the above to be obtained from the office of the Institution,

ALDINE HOUSE, BEDFORD STREET, STRAND, LONDON, W.C. 2.

THE INSTITUTION OF PETROLEUM TECHNOLOGISTS.

The One Hundred and Fifth General Meeting of the Institution of Petroleum Technologists was held at the House of the Royal Society of Arts, John Street, Adelphi, London, W.C. 2, on Tuesday, December 13th, 1927, the chair being taken by Dr. A. E. Dunstan in the absence of the President.

The Secretary read the following list of members who had been elected since the last meeting :—

Members.—Georges Auguste Buire, Thomas William Dagg, Horace Stephen Gibson, Eric Raymond Woodward, Henri van der Waerden.

Associate Members.—Charles Jackson Calderwood, Ernest Lechem.

Transference to Associate Members.—Christian William Breukelman, James Anthony Gerrard McCluskey, George Francis Wallis.

Students.—Frederick George Fortescue, Donald Albert Howes, Patrick Nixon Dick Porter, Leonard Charles Stevens.

Associates.—Alexander Erdely, Benedict William Ginsburg, Frederic I. Lord, Albert Rabson, Herbert Muggleton Stanley.

The Chairman said his first duty was to convey to the members the apologies of the President, who was prevented from attending by important legal business.

It gave him very great pleasure to welcome the lecturer, Mr. H. R. Ricardo, because as everyone knew, his name was a household word in all matters affecting engine design and the very vexed question of detonation. Mr. Ricardo was a very well-known engineer, and it was to be hoped he would tell the Institution all there was to know at the present moment about the design of an engine which would militate against pinking, and that he would describe his own work on that subject. In the course of the discussion, which would be open both to engineers and chemists, something would certainly be said about the chemical side of the subject and information would probably be given as to the various types of mixtures chemists had designed to fight the evil which Mr. Ricardo was going to deal with from the engineering standpoint.

The following paper was then read :—

Mechanical design in relation to Detonation.

By H. R. RICARDO, B.A., M.I.A.E.

I HAVE been asked to discuss the question of detonation in its relation to engine design. In the present state of the art, it is detonation which sets a limit to the compression ratio we can employ and therefore to the power, output and efficiency obtainable. Before discussing any details of design I would like to consider first what could be done in the way of performance if detonation were non-existent.

As we raise the compression ratio of an engine we improve both the power output and efficiency very nearly in proportion to the increase in the air-cycle efficiency and, at the same time, we diminish the flow of waste heat which is the source of most of our troubles in the way of carbonisation, pitted or burnt out exhaust valves, etc. There is still a widespread but entirely erroneous belief that a high compression engine runs hotter than one of low compression—the opposite is of course the case. As we raise the ratio of compression we raise the momentary maximum flame temperature, but only very slightly—in fact by less than 2 per cent. for each whole ratio of compression between, say, 4 and 8 : 1; against this the prolonged expansion reduces the mean temperature during the firing stroke, while the exhaust temperature is of course lowered very considerably, due to the longer expansion, with the net result that the mean cycle temperature is greatly reduced and our weakest link, the exhaust valve, is relieved enormously, since it has to deal with gases at a substantially lower temperature and pressure. This general belief that a high compression engine runs hot is probably founded on two observations :—

(1.) Very high compression engines are apt to be hard on sparking plugs and to suffer from pre-ignition off the sparking plug points. This, however, is almost invariably due to one or other of two causes: persistent detonation which, by re-compression of the already burning gases into the sparking plug recess, causes a severe local temperature rise in the immediate neighbourhood of the electrodes; the other prevalent cause is leakage through the body of the sparking plug—it will be obvious that a very minute leakage of gas at a temperature in the neighbourhood of 2500° C. will suffice to raise enormously the temperature of the thin and heat insulated central electrode.

(2) The other observation is that burnt-out exhaust valves are of fairly common occurrence in racing car engines, which are invariably

of high compression—the explanation here is not far to seek, the racing car engine is usually run at something like three times the speed of its touring brother and therefore, other things being equal, is shipping three times as much heat past its exhaust valves; actually far more than this, for it is running also at a much higher load factor.

It is, in truth, only by using as high a compression as possible that the exhaust valves can live at all in such an engine—a low compression racing engine, were it given sufficient breathing capacity to run at an equally high speed, would burn out all its exhaust valves in the first lap at Brooklands.

I have laboured this point as to the much cooler running of a high compression engine, provided, of course, there is no detonation, for although it is fully realised by all the best designers, the public, as a whole, have an entirely erroneous belief on the subject.

Still, supposing that we are given a fuel which will not detonate under any circumstances, let us consider next what compression ratio we would choose under these happy circumstances. I have said that as we raise the ratio of compression we raise the power output and efficiency more or less proportionately to the increase in the air-cycle efficiency.

The theoretical ideal efficiency at first rises rapidly with increase in compression, but flattens out later on. For example, for a compression ratio of 4 : 1 it is 42·6 per cent., for 5 : 1—47·5 per cent., for 6 : 1—51·2 per cent., for 7 : 1—54 per cent., for 8 : 1—56·5 per cent.

By raising the ratio from 4 to 5 : 1 we gain 11 per cent. in power and efficiency, from 5 to 6 we gain a further 8 per cent., from 6 to 7 a further 5½ per cent., and 7 to 8 only a further 4½ per cent., and so on. On the other side of the picture we must consider the maximum peak pressure which our structure and our moving parts will have to sustain. As we raise the ratio of compression so we raise also the maximum pressure, but in this case we follow a constantly rising, not a flattening, curve. Thus for a compression ratio of 4 : 1 the peak pressure at maximum torque is approximately 360 lb. per sq. in., at a ratio of 5 : 1 it is 490, at 6 : 1—625, at 7 : 1—770 and at 8 : 1—930 lb. per sq. in. Thus between 5 : 1 and 8 : 1 the maximum peak pressure is very nearly doubled, while the ideal efficiency is increased by about 19 per cent.

In any really high speed engine such as an automobile or aeroplane motor, the internal friction of the engine is very largely dependent upon the weight of its reciprocating and rotating members, for it is the inertia and centrifugal forces resulting from these organs which controls the amount of bearing and therefore friction surface required—and here we enter a vicious circle for, as we increase

the bearing surface, of, for example, the connecting rod big-end, so we increase the weight of that member and require yet more surface in consequence thereof. Clearly the weight of such parts as the piston, connecting rod and, to some extent, of the crankshaft is or should be a function of the maximum pressure they will have to withstand; other factors enter in here, however; in the case of the piston, for example, heat dissipation and foundry and machining problems call for certain minimum thicknesses, while the urgent need for stiffness both in the connecting rod and crank set a lower limit to the weight of these parts. So far as relatively low ratios of compression are concerned, the minimum weight compatible with heat dissipation, stiffness, and practical foundry and machining conditions will afford ample strength to resist the peak pressure, but when we come to consider compression ratios of the order of 8 : 1 and upwards this is no longer the case, and we then find that the additional weight necessary to enable these parts to sustain the higher pressures will involve additional friction to an extent which may easily absorb the whole of the gain in efficiency resulting from the higher compression; this, as we have seen, amounts to a bare 4½ per cent. between 7 : 1 and 8 : 1—nor is this the only consideration for, in the case of automobile engines, the mean load factor is only about one-third, hence an increase in internal friction amounting to 2 per cent. at full load becomes 6 per cent. at the normal running condition.

As the size of the engine decreases so the relative strength of the organs increases, for we are bound throughout by a certain minimum thickness governed by commercial foundry and machine shop limitations.

As a broad generalisation, one can say fairly safely that it will not pay, at all events for automobile engines, to employ a higher compression when to do so necessitates adding any extra weight to the reciprocating parts.

Yet, again, there is another consideration to be taken into account, namely, that as we increase the ratio of compression so the volume of the combustion space is reduced while its area, which is controlled by the accommodation of the valves, remains practically the same. Although the actual heat loss to the surface of the cylinder head during combustion is normally very small, yet the loss due to the delayed and incomplete combustion of the stagnant layer of gas adhering to this surface is very serious and this, of course, increases in proportion as the ratio of surface to volume is increased, that is, as the compression ratio is raised. It is probably true that at the higher densities the layer of lost gas becomes somewhat thinner, but not in proportion, and so to our

already flattening efficiency curve is added a rising loss due to the increasing proportions of the surface layer.

Under the average manufacturing conditions of to-day, taking all the above factors into account and assuming that we have a fuel which will not detonate under any circumstances, I think we should compromise on the following compression ratios; for cylinders of 2 to 2½in. bore, 8 : 1, 2½ to 3in. bore, 7·5 : 1, 3 to 4in., 7 : 1, 4 to 5½ in., 6·5 : 1.

In the case of racing car or aeroplane engines one can afford to go to higher ratios because, on the one hand, they are always working at much higher load factors, and on the other, we can afford to spend more time and money on the removal of superfluous weight and on the use of fancy materials.

Up to the present I have been considering what we should do if detonation did not intervene to prevent us, and I have endeavoured to show that we should at once go to much higher compression ratios, and should probably strike a balance such as I have suggested above.

The phenomena of detonation in relation to the chemical compositions of the fuel have been and will be discussed by chemists and physicists far more competent than I to deal with the subject, and I will therefore confine myself to its relation to engine design. So far as my investigations go, the tendency to detonate appears to depend upon a large number of design factors, which I have endeavoured to set down, as far as possible, in the order of their importance.

(1) The length of flame travel from the sparking plug to the farthest point in the combustion chamber.

(2) The degree and nature of movement of the unburnt gases within the combustion chamber.

(3) The temperature and density of the gases at the time of ignition.

(4) The mean surface temperature of the combustion chamber, and more particularly of that part of the surface most remote from the sparking plug.

In order to meet the first condition we must make the combustion chamber as compact as possible with the sparking plug as nearly as practicable in the centre, not quite in the centre, because one portion of the surface; that including the exhaust valve head, is always much hotter than any other, and in order to prevent detonation taking place in this zone it is desirable to offset the sparking plug slightly towards the head of the exhaust valve, and so shorten the flame travel in this particular direction. From the point of view of flame travel, the most efficient head we can in practice

employ is that of the conical type with inclined overhead valves and with the sparking in the centre or just slightly off centre. In order, however, to get the sparking plug in this position we have to use two cam-shafts, so that the engine becomes costly, complicated and somewhat heavy. This is the form used on practically every successful racing car at the present time. When, owing to the use of a single overhead cam-shaft, the central position of the sparking plug is forbidden, we can compromise by using two sparking plugs accurately synchronised, one on either side of the combustion chamber, though not necessarily out at the extreme radius. In this case we find it necessary to give a slight lead usually 3 or 4 degrees to the ignition on the exhaust side, in order to ensure that the flame travel shall not finish up against the exhaust valve head. With this arrangement the result is very nearly equal as regards performance and freedom from detonation to that obtained with the single central plug, but it is then dependent on the functioning of both sparking plugs and their accurate timing; if either plug fails the performance falls off heavily and the tendency to detonate is greatly increased, more especially so when the plug on the exhaust valve side cuts out. This form of combustion is used in nearly every aero-engine of to-day, in preference to the single central plug, the argument being that, an aero engine must have duplicate ignition system, on the grounds of reliability, and this being the case, the additional weight of an extra overhead cam-shaft, to allow of the central plug position, cannot be tolerated. If either ignition system fails, although the performance of the machine is at once greatly reduced, it can still continue in the air.

The type of combustion chamber containing two overhead vertical valves operated by push rods is attractive superficially, because the chamber appears very compact, is truly cylindrical, and can be machined all over. The fly in the ointment is that the two overhead valves compel the banishment of the sparking plug to the extreme side and therefore involve a very long flame, run right across the full diameter. With two ignition systems accurately timed, as in an aero-engine, this would give a very good performance, but the average motorist would not tolerate the nuisance of maintaining two sets of ignition in perfect synchronism. With a single sparking plug only, its performance is rather poor, not only is the tendency to detonate very acute on account of the long flame travel, but owing to poor turbulence the spread of flame is much too slow, necessitating constant spark adjustment with change of speed or throttle opening. We have experimented a good deal with all three types of combustion chamber, and find that their relative merits, as regards tendency to detonate, are roughly as follows—I say roughly advisedly, for although we have done our

very best to make the tests truly comparative it is quite impossible to maintain all the relevant conditions such as turbulence, etc. constant throughout:—

Inclined overhead valves central sparking plug H.U.C. 6 : 1.
Inclined overhead valves two ignition points H.U.C. 5·9 : 1.
Inclined overhead valves plug on exhaust side only 5·6 : 1.
Inclined overhead valves plug on inlet side only H.U.C. 5·3 : 1.
Vertical overhead valves single plug radially H.U.C., 5·2 : 1.

In the case of a side valve engine one is confronted at once with the difficulty that the distance from the outer edge of the exhaust valve to the farthest point in the cylinder bore is necessarily a very long one, and even with the plug in a central position the minimum flame travel is still almost as long as in the case of the overhead valve engine with the plug at the side—moreover, owing to the effect of the valve pockets, the initial turbulence is very much damped down, the area of the stagnant layer of gas is very large and there is insufficient turbulence to sweep it away. We found, however, some ten years ago, that by concentrating the combustion chamber as much as possible over the valves and bringing the remainder of the head down into the closest possible contact with the piston at top dead centre, quite a compact chamber could be obtained, for the thin film of gas entrapped between the piston and flat part of the cylinder was so chilled by the two surfaces as to prevent any possibility of detonation in that zone—in other words, this portion of the combustion chamber could then be put completely out of action; moreover, by somewhat restricting the passage between the effective combustion chamber and the cylinder, any desired degree of turbulence could be produced during the compression stroke. Under these circumstances the combustion chamber becomes roughly triangular in plan, with one apex over each valve and one over the piston, while the plug is approximately in the centre. With such an arrangement the maximum length of flame travel is reduced to but little more than the radius of the cylinder (depending to some extent on the size and positioning of the valves) and the combustion space is nearly similar to that of the two cam-shaft overhead valve type, but displaced to one side instead of being vertically over the cylinder bore. So far as detonation is concerned it is comparable with the two cam-shaft variety and, of course, having a centrally placed plug, single ignition suffices. Following the same comparison as I gave previously and taking the H.U.C. of the central plug overhead valve type as 6 : 1, that of the side valve type may range from 5·4 to 6 : 1, depending on the degree of turbulence provided and upon the spacing of the valves, while that of what I might

term a conventional side valve head is in the neighbourhood of 4·3 to 4·6 : 1.

I have emphasised that in order to render the film of gas between the piston and cylinder head inoperative as regards detonation the space must be kept very small. I have lately seen a number of so-called semi-turbulent combustion chambers in which the space allowed above the piston is far too great; such heads show a slight improvement over the conventional flat or sloping type owing to the greater turbulence, but no improvement as regards detonation, since this can and does occur in the space between the piston and cylinder head. I mention this point because again and again I have had such examples brought before me to illustrate that there is little or no difference as regards detonation between such a turbulent head and the conventional type—yet others have had a close clearance but a narrow passage extending right across the piston, thus restoring again the full length of flame travel and so completely destroying its advantage as regards detonation.

The worst combustion chamber I have yet met was one of the T head type with inlet and exhaust valves in shallow pockets on either side of the cylinder, and with the sparking plug fitted in the inlet valve cap—this had a flame travel equal to rather more than twice the cylinder diameter and finishing up in the hot exhaust valve pocket. As might be expected it detonated heavily and had an H.U.C. of only 3·6 : 1.

Turbulence.

We come next to the question of the influence of turbulence upon detonation. It is, of course, well known that we depend entirely on the mechanical movement of the gases within the cylinder to speed up the combustion process which otherwise would be altogether too slow even for the slowest running engine. By varying the degree of turbulence we can control, at will, the speed of combustion. It is, of course, fairly obvious that we must have extremely rapid burning, for we do not want to start it appreciably before the piston has reached top centre, and equally we want to complete it before the piston has travelled appreciably on its outward stroke, for obviously we want all the expansion we can get, not only on the grounds of efficiency alone, but even more so for the sake of our exhaust valves. Delayed burning means, of course, higher exhaust temperatures and pitted or burnt exhaust valves. I spoke also of the thin layer of gas always adhering closely to the cool walls of the combustion chamber and therefore so chilled as to escape complete combustion; here again turbulence is the remedy in order mechanically to scour away this layer, just

as, in the converse case, one stirs a saucepan to avoid local scorching. Finally, we rely on turbulence both to prevent any portion of the yet unburnt gas from being cornered and detonated by the oncoming flame front, and to break up that flame front into skirmishing parties. At the other end of the scale, excessive turbulence is also objectionable in that by scrubbing against the cold walls the loss of heat is greatly increased, while from a mechnical standpoint too rapid combustion and too rapid a rise of pressure causes severe shock and, in extreme cases, gives rise to an actual knock almost resembling detonation. While there is little doubt but that turbulence per se tends to reduce detonation, yet it is extremely difficult to arrive at any quantitative relation between the degree of turbulence and the tendency to detonate, because any variation in turbulence produces so many secondary effects which all have an indirect influence on detonation; for example, increase of turbulence involves a decrease in the temperature of the exhaust valve head, and we know that the temperature of the exhaust valve is far above the ignition point of the fuel and high enough to produce local surface combustion during the compression stroke, thus it has in itself a powerful influence on the tendency to detonate.

Yet again, it is difficult to vary the degree of turbulence without varying also some other relevant factor such as the temperature of compression, or even the length of flame travel. Further, mere turbulence is by no means the whole story, for so much depends upon the nature of the movement of the gases. As an experiment we have tried in an engine directing the turbulence to take the form of an organised rotational swirl at varying speed, measured by an anomometer vane within the cylinder; even when the gases are rotating within the combustion chamber at the enormous speed of 15,000 r.p.m. the effect, so far as rapidity of combustion is concerned, is that of an almost stagnant charge, for burning continues slowly throughout the whole of the expansion and is not always completed even at the end of the exhaust stroke, with the result that the next charge is ignited on entry by the lingering flame from the previous cycle. Presumably in such a case the nucleus of flame initiated at the plug points is merely whirled round, and is occupied in chasing its own tail rather than in spreading out to new fields of adventure. As might be expected, however, the H.U.C. is then very high. Again, turbulence may take the form of mere local eddies as distinct from the general rough and tumble which we are aiming for. On one experimental side valve engine designed purely for research in combustion chamber form, we have tried some 45 different forms of combustion chamber giving varying degrees of turbulence, varying lengths of flame travel, etc., and the highest useful compression consistent with freedom from detonation has

ranged, on the same fuel, from 5·0 : 1 to 6·6 : 1, though some of these —the best, so far as detonation is concerned—are hardly practical on account of excessive turbulence.

The best practical form we have yet arrived at will stand a compression ratio of slightly over 6·0 : 1.

Effect of Cylinder Size on Detonation.

We have lately had good opportunity for observing the effect of cylinder size on detonation on a range of single cylinder sleeve valve engines of 2¾, 3½, 5½ and 8½ in. bore respectively—all have very nearly the same stroke/bore ratio, the same form of combustion chamber, namely, a shallow cone with the sparking plug in the centre at the apex, and in fact are as nearly similar as is possible over so large a range of size. In each case also the maximum length of flame travel was approximately 25 per cent. greater than the cylinder radius. The highest useful compression ratio as determined for each of these engines on the same fuel at the same working temperatures and at the speed corresponding to maximum torque in each case was found to be as follows: 2¾in., 7·9 : 1; 3½in., 7·3 : 1; 5½in., 6·2 : 1; 8½in., 5·4 : 1. These observations, when plotted, will be found to lie on a reasonably smooth curve, except that the 5½in. cylinder is rather low, due probably to some subtle difference in the degree or nature of turbulence. It will be observed that the cases of sleeve valve engines I have quoted are capable of operating at a much higher compression ratio than the poppet valve examples I referred to previously. This is due, in part, to the fact that there is no hot exhaust valve present in the combustion chamber, in part to the fact that the absence of valves permits of a more compact and symmetrical head than is possible even with the inclined overhead valve poppet engine; and, of course, a central sparking plug can be used to the full advantage. In all the various comparisons I have made throughout this Paper the same fuel was used in all cases, and where any difference in size actually existed the figures I have quoted are all reduced to terms of cylinders of about 4½in. bore—on the basis of the sleeve valve experiments I have just referred to.

DISCUSSION.

The Chairman (Dr. A. E. Dunstan) said the paper was to have been one of a series of two, the one dealing with the matter from the engineering point of view and the other from the chemical. The crowded state of the Institution's programme had made that impossible, and the result was that that evening the discussion would be thrown open both to chemists and engineers. There were so

many guests present who would wish to take part in the discussion that he was reluctant to interpose any remarks of his own, but there was one point he wished to raise. Although the author had provided a standard engine, so far the chemists had not decided on a standard fuel. It was a most difficult thing to try to piece together the evidence one found in the technical journals concerning the compression ratios of different fuels, when each observer was talking about a different engine and a different fuel. It might be possible to use a mixture of octane and heptane, as Dr. Edgar had proposed, and decide to standardise that as the fuel for work on engine design. He hoped the point would be taken up and developed during the discussion.

He would ask Dr. Ormandy to deal with the chemical side of the question.

Dr. W. R. Ormandy thought the Institution was very much to be congratulated on having Mr. Ricardo to deal with the engineering side of the question, but it made it very difficult for whoever was to follow on the chemical side. Mr. Ricardo held the Medal of the Institution of Automobile Engineers and only this month had received the Crompton Medal.

The question of fuel and engine became more and more important as time went on. In the old days, when petrol consisted of the light fractions, things were comparatively easy, because engines had a low compression, and it was a well known fact that the tendency to detonate in any range of fuel increased as the molecular weight increased. The heavier oils, such as kerosine, tended to detonate much more than aviation spirit. As time went on and requirements could not be met with the light spirit, heavier spirits were used, and he thought the first stage was the discovery that it was desirable to add benzol to a fuel, because the addition of benzol gave it a higher resistance to detonation and enabled it to stand a higher compression. Then came the celebrated work carried out at the instigation of the Shell Oil Company by Mr. Ricardo on the relation of fuel constituents in general to the compressions that they would stand, and the programme that was then set by the Shell Company of selling a fuel which had a standardised compression ratio when measured on a certain engine.

However, the demand grew and he thought it became impossible to keep to that standard, because the world supply of suitable high compression spirit, rich in aromatics and napthenes, was insufficient. The quality of the fuel would undoubtedly have gone down, in his opinion, but for the recourse which had to be made to the cracked spirit. There was no doubt that cracked spirits, which were as variable in their nature as the natural spirits, were, on the

whole, better than the average straight run spirit in their ability to stand compression.

Wonderfully comprehensive as was the work that Ricardo did on behalf of the various constituents of the average fuels, he did not touch on, or only touched on very superficially, the question of the behaviour of the engine in relation to unsaturateds. He was glad to think there was now a prospect of that work being taken up. Certain funds were available, and his brother chemists might like to know that not the least part of the difficulty was to determine what was an unsaturated. It was a matter which had not been altogether settled, but there was a prospect that experiments would be carried out by Ricardo shortly on pure olefines, which would at any rate give something to go on. What was quite certain was that cracked spirit could stand a very high compression ratio. He believed a Russian crude cracked on the Cross plant had a compression ratio of 5·9 on the standard engine, which was quite a high figure.

Though the chemist had kept saving the situation by various means, such as cracking, he had not been idle in other directions, and had laid himself out to find materials which had a very high effect in reducing the detonating properties of a fuel when added in small quantities. The most brilliant example of that was lead-tetraethyl.

It was very interesting to hear both sides of the question. The chemist had made many attempts to produce a better fuel, but his efforts were conditioned by the enormous quantity of material he had to provide, so that whatever change was brought about must be something which was capable of being carried out on an enormous scale. On the other hand, the engineer had done what he could to render the use of fuel of such a high grade in that respect unnecessary. In view of the figures given in the paper as to compression ratios which the author apparently considered "quite reasonable" with a properly designed engine, one could only be struck by the tremendous quantity of badly designed engines on the market to-day, because the figures given by the author would demoralise the bulk of the engine makers. He talked of a compression of 6 as being quite reasonable on a 4-inch cylinder, whereas personally he thought the average 4-inch cylinder was quite content with something very much nearer 5, and there would be trouble even then unless care was taken as to the fuel used.

He proposed to leave the question of the positive value or otherwise of such dopes as lead tetraethyl to other speakers. Personally, he should not like the idea of having two sparking plugs which had to be synchronised, but he would not object to a two-pole plug which he could put in series with a second plug, and he would like

to know whether it was possible to design a two-pole plug with electrodes heavy enough not to cause pre-ignition, and sufficient insulation between them to prevent short circuiting in the space allowed, and also whether it was possible to get the compressions which the author had referred to as being "reasonable" without at the same time having to use such turbulence that the pressure increment per degree of crank shaft rotation was not so high that uneasy running was produced unless the engine was designed with such a margin of strength and stiffness in the crank-case and connecting rods as to cause it to cost a good deal, or to be unduly heavy. Unfortunately, nearly all the cures for one evil brought other evils in their train, and adequate turbulence to bring about the ability to use a low-grade fuel at a high compression might be accompanied by other evils which were perhaps as difficult or so costly to overcome that they were hardly within the region of practical politics.

Mr. A. C. Egerton said he had been very interested to hear what Mr. Ricardo had to say on the engineering side of the interesting problem of detonation. Dr. Ormandy had referred to the difficulty of saying whether a substance was unsaturated or saturated. There had also been some difficulty in determining whether a thing was organic or inorganic!—but the great difficulty in the present case was to say whether a thing was detonating or not detonating. Personally, he did not quite like the word "detonation" in that connection, because he did not think the phenomenon under consideration was a true gaseous detonation. If it were a true gaseous detonation he did not think it would be possible to travel in a motor car. He had made a great many experiments on the detonation of gases, and the effect on the steel tubes he used was that such an amount of the iron itself became shattered and fused that he did not think an engine would stand it. Furthermore, he had not been able to get any true detonation of gases in a mixture of gases equivalent to the mixtures used in engines; *i.e.*, with pentane and hexane.

One did get in the course of such investigations, however, what he might describe as "pseudo-detonation": *i.e.*, a new detoning type of combustion suddenly starting but not continuing.* Whether engine knock was that type of detonation (he had only got it in fairly strong mixtures) he could not say, but he was inclined to think it was more of the vibratory type which one got when an explosion became audible. In the case of an audible explosion there was always the effect of vibration set up, and he thought the knock was only an advanced form of that vibratory type of explosion.

* Proc. Roy. Soc., 1927, A. 114, p. 141.

Others might take a different view, but as a matter of fact that opinion was not very different from regarding it as a sort of pseudo-detonation, because that vibratory type assumed that in a certain portion of the charge there was an accelerated combustion, which was then reduced owing to a change in the density of the gas due to the vibration set up.

As a matter of fact, that view fitted in rather well with the effect of anti-knocks, because anti-knocks had no effect on a rapid, normal type of explosion such as one got with detonation. If one had a combustion which was going to become a gaseous detonation, the rate of that combustion did not seem to be altered at all by the presence of anti-knocks. If, on the other hand, one was going to have a vibratory type of explosion, the anti-knock was decomposed during the first movement of the head of the flame, and was then broken up and ready to act. He had shown that anti-knocks under those conditions affected the rate of flame travel where there was time for the anti-knock to be broken up in front of the flame.

He did not altogether like the use of the word "detonation" in the present connection. If the phenomenon was regarded as auto-ignition he did not know whether that would help, because an auto-ignition was an ignition in front of the flame travel, and if it was in the nature of a detonation, that is to say, an activation of a proportion of the charge which meant that the whole of the gas in the neighbourhood was going to ignite, it was a detonation, which brought one back where one was to start with ; whereas if it were a simple ignition it would accelerate in the normal way, as would occur with multiple ignition, and that tended to allay detonation rather than promote it.

Dr. G. B. Maxwell said there were one or two questions he wished to put to Mr. Ricardo, and also one or two criticisms he wished to put forward. He spoke purely as a layman in engine design, but as a chemist who had been working on flame propagation and to some extent on the theoretical aspects of pinking. In the first place, he wished to add his protest to Mr. Egerton's to the use of the word "detonation" to express what he thought in England was more aptly termed pinking. In his own work on the effect of compression on the inflammation of paraffin hydrocarbons under conditions which approximated to those of an engine he was able to obtain effects which certainly would give the effect of pinking in an engine, and to photograph those effects, but he had never found anything similar to true detonation. Such effects appeared to take place in the end products of the flame, which he thought rather fitted in with some work Mr. Ricardo referred to on the use of a stroboscope by which pinking was observed to occur very late in

flame travel. Like Mr. Egerton, he got a very highly vibratory flame, and the moment it was checked by the end of the cylinder a pressure wave was produced and a glow of illumination spread throughout the contents of the cylinder. The most powerful effect was produced at the ends, and especially near the sparking plug, which might tend to account for the fact that pinking might lead to pre-ignition from the plug points.

With regard to the point Mr. Ricardo had raised of the influence of compression ratio on the efficiency of an engine, in the earlier Papers by Tizard and Pye in the Empire Motor Fuels Report those authors stated they had found the increase was more than proportional to the increase in the air standard efficiency, and that was rather what he would expect from his own work. He had done some work on the effect of pressure on the rate of flame travel and on the pressures produced in paraffin-air mixtures, and he found he got the maximum pressure and flame-speed with a rather weaker mixture if he raised the initial compression. He presumed that this was simply an effect of repressed dissociation in the normal manner which naturally would not be so marked in an engine owing to association during the expansion stroke, but he would like to know whether Mr. Ricardo had not found some increase over what he would expect from pure proportionality to the air standard efficiency.

On the question of turbulence, he thought that in the early days Mr. Ricardo was under the impression that turbulence did not really of itself affect the tendency of a fuel to pink, and he wondered whether he had had further evidence which had caused him to change his mind. In Mr. Ricardo's work on the high-speed internal-combustion engine there was a direct statement that turbulence did not of itself affect the tendency to pinking.

In dealing with the experiments on the effect of cylinder bore on the tendency to pink, the statement was made that in each case the speed was such that the engine was giving the maximum torque. He presumed that if the piston speeds were the same in each case and the other factors were in proportion the number of revolutions per minute of the widest bored engine would be less than that of the smallest bored engine. In other words, there was a variable factor coming in there which was known to affect pinking. He wished to ask Mr. Ricardo whether he thought that the effect of the drop in speed as the cylinder bore increased was not possibly a contributory cause of the lowering of the compression ratio. Personally, he had not found that the length of flame travel in a closed cylinder had such a profound effect on the tendency to pink—assuming that the effects he had noticed were pinking. Two effects came in in the experiments Mr. Ricardo had made: (i.) the slower speed of the

engines of wider bore, and (ii.) the possibly greater temperature of the centre of the piston head. The effect of hot spots was too well known to need comment.

Mr. E. R. Redgrove congratulated Mr. Ricardo on his excellent Paper. He found the pressures given for the various compression ratios most interesting, because they showed that without pinking at a compression ratio of even 8 to 1 one had to deal with only 930 lb. per square inch. He did not think any respectable lubricating oil would be troubled by a pressure of that magnitude. The work which had been done and which was now going on tended to show that boundary conditions of lubrication would not be reached until pressures five or six times greater than that were attained. The pressures given by Mr. Ricardo, of course, referred only to pressures obtained without pinking, or that was what he understood. If serious pinking occurred it was quite possible that those pressures would be increased three or four times, and therefore it became a question not only for the engineer so to design his engine that pinking did not occur and for the fuel chemist to get a fuel that would stop pinking, but also for the lubrication chemist to see that he could provide a lubricant which would safely take care of the bearings should pinking occur, even with a compression ratio of 8 to 1.

Dr. W. R. Ormandy, referring to the question of a standard fuel, said the problem was a serious one. Mr. Ricardo reported the compression ratios on his engines carried out with the same fuel, but one did not know what fuel was used, and even if one did it might not be possible for anyone else to get hold of exactly the same fuel. Numbers of experimental engines were in use and a good deal of work was being done on them, but to a very considerable extent that work was wasted from the point of view of co-operative research, because one could not compare one compression ratio with another on two different engines with different fuels. He believed standard octane was being used in America in considerable quantities, but for purposes of comparison it would be necessary to get the same octane, and there were quite a number of octanes, as the formula showed. He had been told by the chief research chemist of the Gasolene Corporation that they had, at very great expense, produced some very small quantities of the whole eight, and the difference between one octane and another was greater than the difference between octane and benzol.

Fortunately, it was possible to get one single hydrocarbon, heptane, in a pure state, and that was possible not because it was produced by petroleum technologists but because it came from the leaves of a tree. It was the only hydrocarbon which was what

might be called respectably pure. Any suggestions from those taking part in the discussion of materials which could be used would be very valuable if they could be carried into effect. One might consider using, for example, heptane and benzol, but they would both stand a comparatively high compression, and what was wanted was something of the requisite volatility and pure and having a boiling point of 80° to 110° or possibly 120°, which would have a really bad compression ratio, so that any desired mixture could be obtained for purposes of experiment.

Mr. E. C. Craven said he wished to ask Mr. Ricardo if more than two sparking plugs had ever been tried and if in fact multiple ignition did tend to reduce detonation. On the one hand, dealing with chemical theories, and anti-knocks and the like, there was the theory that lead tetraethyl fired the mixture at a huge number of points throughout the combustion space, so that the flame travel was greatly reduced and the tendency to detonation thereby reduced also. On the other hand there was the latest theory of Callender, where ignition is supposed to occur in a rather different way, but also at a large number of points distributed throughout the combustion space, and it was stated that that actually gave rise to detonation. There was a direct contradiction there, and he would like to hear the author's opinion on it, and on the effect of multiple ignition generally, especially when it was extended to a large number of points.

He did not know how the nature and degree of turbulence were measured or could be estimated, and would welcome any information on that subject. He would also like to know whether any stirring device could be introduced into the cylinder of an engine, and what would happen if a handful of steel balls was put into an engine.

Mr. C. de Ganahl wished to ask a question with regard to the anti-knock figure obtained on different fuels with Mr. Ricardo's engine. For instance, suppose a certain standard of test was used whereby so many watts continuously heated the intake air, and the air fuel mixture was adjusted to maximum power output, and the H.U.C.R. reading then taken when the engine was detonating on every stroke. Then on testing any fuels, A, B and C, H.U.C.R. readings of 5·1, 5·2, and 5·3 respectively were obtained. Now suppose the method of test were changed, i.e., the electric heating of the intake air was reduced by one half and air fuel mixture other than that of maximum power were used, and the H.U.C.R. reading taken at the beginning of detonation.

He realised that the readings obtained would be radically different, but could a sequence of the readings or the relative value of the readings be changed?

Mr. Harold Moore wished to ask one or two questions connected with engine design, or rather the effect of engine design. He wished in the first place to know whether any figures had appeared giving the relative merits of an engine with a single valve and distributing valve—a single valve operating as both inlet and exhaust valves and a distributing valve in series behind. That was a design which had been patented innumerable times and one which appeared to overcome some of the difficulties in connection with detonation.

Another type which would be of interest was the Junker type of engine, with two opposed pistons and no cylinder head, and which gave room for any degree of turbulence by raking both inlet and exhaust ports, thus giving a rotary motion to the gases; this design also had the advantage of giving a very simple method of assisting scavenging due to the uniflow action of the gases.

There was reason to suspect that the unsaturateds—they were all classified roughly as unsaturateds—generated from a Pennsylvania base or a paraffin base oil by the cracking process had quite a different anti-detonating effect from those generated by the cracking of the naphthene base product. Chemists were accustomed to lump all the unsaturateds together, but an unsaturated product generated by cracking a naphthene might still be a naphthene as well as an olefine. Some figures he had had before him of practical results on the products of cracking seemed to indicate that though when one took a naphthene base crude and cracked it one obtained a very high anti-detonating value for the fuel, the corresponding product from a paraffin base crude did not give anything like the same degree of enhanced anti-detonating properties. Perhaps some member of the audience could give information on the point.

Mr. E. S. Beale asked if the author could give any explanation in detail of the observation which had often been made of the difference in the smoothness of running between an engine running, say, on coal gas and an engine running on liquid fuel such as petrol. It had been suggested that the difference lay chiefly in the degree of mixing of the fuel and the air, and if that was the case it seemed possible that a bad distribution of fuel and air in the case of a petrol engine could exist in different parts of the same cylinder. If that was carried far enough there would be a distribution so bad that the liquid was actually concentrated in drops and therefore as such could not burn; and that reduced to Dr. Callender's nuclear theory.

He had recently made an observation which might suggest another mechanism by which turbulence in a mixture would effect the

degree of detonation under given conditions. The nuclear theory of detonation depended entirely on whether a fuel mixture would condense on compression or, in the other direction, if on compression it would be evaporated. In that respect hydrocarbons differed from the usual familiar substances such as water, but he thought the vapour of hydrocarbon when unmixed with permanent gas would condense on compression. When mixed with air in the same proportions as existed in the ordinary combustion engine it did exactly the opposite, and in that connection it seemed possible that the degree of turbulence of the mixture would decide what mixture strength existed in the neighbourhood of the drop or where the drop was before it completely evaporated. If the mixture strength was very high in this region and the composition of the mixture at that point was practically pure hydrocarbon vapour it would recondense, but if by the time the ignition occurred the turbulence had mixed that vapour with the surrounding air to a sufficient extent it would not condense on compression.

Mr. A. Rabson said Mr. Ricardo had shown that the position of the sparking plug in the combustion chamber had a great effect on detonation, but personally he would like to know about the position of the spark itself, whether the plug projected into the chamber or whether the spark was flush with the bore or pocketed. The author spoke of a standard fuel, and he would like to know whether the mixture used was constant; he did not know what fuel was used.

Mr. Ricardo asked if the mixture ratio of the fuel itself was meant.

Mr. Rabson said that was what he had in mind, and what the effect of weaker and stronger mixtures was on detonation.

Mr. A. C. Egerton said the author mentioned that in the case of a narrow exposed chamber it was the cooling of the walls which reduced the tendency to detonate. Personally, he was inclined to wonder whether it was not due to the extended surface. He was rather interested in the question of the effect of surface in the preliminary stages of combustion and thought it would be valuable to know whether the author had observed that extent of surface tended to reduce detonation; i.e., to prevent knocking or whether it was only the effect of cooling.

Mr. Ricardo, in reply, said Dr. Ormandy had made some very kind remarks, which he appreciated, and had referred to the fact that no tests on unsaturated fuels had been published. There were two reasons for that. Numbers of tests were done, but they were not published, in the first place because in most

cases with unsaturated fuels the detonation point was remarkably difficult to determine. They showed a wide range of compression ratios, unlike most others, over which detonation was sporadic, and it was therefore most difficult to determine with any approach to precision. In the second place, between themselves they were very erratic. He had a little bone to pick with chemists there, because whenever the samples were returned for re-examination they were found to be entirely different.

Lately he had been carrying out a long series of tests with ethylene. It had a peculiar effect; it would stand an exceedingly high compression ratio for an exceedingly short time, and it then developed pre-ignition without any detonation at all. Water cooling of the exhaust valves and every possible way of cooling the plug had been tried without result, nor had it been possible to find any compression ratio at which it would run continuously without that developing pre-ignition sooner or later. At 7 to 1 it would run for 2 minutes—long enough to get quite a good observation of efficiency—and then break down into pre-ignition. At 3·8 to 1—the lowest figure to which it was possible to get—after half an hour pre-ignition supervened, and therefore he hesitated to quote any figures for the ethylene, for example.

Dr. Ormandy and several other speakers raised the question of what fuel all the compression ratios related to. He ought to have made that clear in his paper. They all referred to standard Shell Aero Spirit. That was the standard Air Ministry specification as supplied by the Shell people.

Dr. Ormandy: Which never varies?

Mr. Ricardo said it did not vary a very great deal, at any rate not sufficiently to affect the relative value of the observations by more than 0·1 of a ratio.

With regard to two-pole sparking plugs, he had often tried to get the sparking plug makers to produce a two-pole plug, but had not so far seen one which was at all satisfactory. The two-pole plug did not wholly meet the needs of the case, because one wanted to be able to spark a little ahead on one side of the cylinder, and the two-pole plug would only do it simultaneously.

So far as compression ratio was concerned, he had quoted a series of experiments on cylinder heads on the side valve engine, and pointed out that the highest useful compressions ranged from 5 to 1 to 6·6 to 1, and that the highest found really practicable was 6·1. That gave a pressure rise of 35 lb. per sq. in. per degree crank angle, which in a reasonably well designed engine was quite tolerable. At 6·6 to 1 there was a pressure rise of 55 lbs. per degree, and he would not care to drive that. He thought,

however, that the figure of 6 to 1 was quite reasonable as applied to any normally well designed engine.

Dr. Egerton raised the point of whether the freedom from detonation between the two closed surfaces was due to surface or cooling. Personally, he was afraid he had no means of knowing to which it was due. He had watched the spread of the flame, across the combustion chamber with the stroboscope. One saw the flame from the plug rapidly accelerating until it reached the narrow gate into this thin film and then it rapidly slowed down again, and as the depth of the film was increased so was the slowing down lessened, until at round about ¼ in. deep it appeared not to slow down at all; it passed straight across. Whether that was a cooling effect or a surface effect he had no means of knowing.

Dr. Maxwell first raised the question of the definition of detonation. Dr. Maxwell was much more of a chemist and physicist than himself, and personally he had simply made use of a term which had been coined. He thought "pinking" was probably a better expression. In any case it was less controversial.

With regard to the increase in efficiency with increase in compression ratio, if one took the case of the variable compression engine, which was a highly efficient engine with a very long stroke, one did observe an increase of efficiency with an increase of compression, but he had been thinking rather of the case of the more normal commercial engine, which usually had a much smaller stroke-bore ratio and a more straggled-out combustion chamber. Generally speaking, in those cases a high compression meant a much greater proportion of surface to volume, and consequently as the compression was raised there was a slight drop in the relative efficiency.

So far as the question of variation in speed on the various engines he had mentioned was concerned, it was very difficult to decide on what basis to compare engines of widely different size. Generally speaking they were all compared, for the purposes of the present paper, on the speed at which they developed maximum torque, which was very nearly a constant piston speed in all cases. He had noticed, however, in the case of the variable compression engine he used that the highest useful compression ratio did not vary very much with speed, except at very low speeds, when it became markedly greater due, he thought, to the effect of surface combustion off the exhaust valves. Between 900 and 1,800 revs. there was very little difference in the highest useful compression. Actually the point of maximum torque was practically the point of maximum pinking. The same would apply in the case of the sleeve valve engines.

Dr. Maxwell doubted whether flame travel had so important an effect as the paper seemed to indicate. Personally, he had become

more impressed with the effect of flame travel from the experiments he quoted on a side valve research engine. There one found that flame travel, in that engine at all events, appeared to be the governing factor all the time, and watching the flame through the stroboscope, one saw the increased velocity and could spot the pinking starting from the far corner.

It was quite true that some years ago he thought that pinking had nothing to do with turbulence, and that there was no particular relationship between them, and even to-day he did not feel at all sure about it, because it was so impossible to vary the turbulence without varying so many other factors. He merely gave the figures for what they were worth. Generally speaking, he thought there was a fairly strong reason for supposing that increase of turbulence reduced the tendency to pinking, but even that wanted qualifying, because it depended a good deal on the nature of the turbulence. He would not care to be categorical about it.

Mr. Redgrove referred to the pressures used and their effect on lubrication, and pointed out that with a compression ratio of 8 to 1 and the figure given in the paper of 930 lb. to the square inch on the piston there would be no trouble with any reasonable lubricant. Personally, he had never supposed that in this matter the lubrication was in any way the limitation; he did not think there was any possibility of difficulty in that direction, but at the same time with a pressure of 930 lb. per square inch on the head of the piston the pressure on the connecting rod big end bearings and on the gudgeon pin bearing was enormously greater. As a matter of fact, he was working quite comfortably with gudgeon pin pressures of well over 6,000 lb. to the square inch, and having not the slightest trouble with lubrication.

He could find no evidence at all that there was any general pressure rise of the cylinder when pinking occurred, nor did he think it possible that there could be, because there was not the heat evolved in the mixture to produce any higher pressure than the normal working pressure.

Mr. Craven asked about experiments with multiple ignition. At one time he ran a variable compression engine with four point ignition round the cylinder. It was given up because it was difficult to ensure that all four points were always sparking, and the general maintenance was too troublesome. He then dropped to two points on opposite sides of the cylinder, and in doing so altered the H.U.C. value, as near as could be judged, from 0·05 to 0·1 of a ratio. If one dropped to one point the H.U.C. value dropped a further 0·4 to 0·5 of the ratio. He thought that was reasonably consistent with the question of flame travel, because there was no very great

difference in the flame travel between two opposite points and four.

Mr. Craven also asked how turbulence was measured. The answer was, empirically. The rate of pressure rise after combustion had fully started was regarded as measure of turbulence. One got at first a gradual rise up from the compression curve and then a perfectly straight rise of pressure, as shown by the indicator diagram, and it was judged entirely by the slope of that line. Various stirring devices had been tried, not actually in a working engine but in a single cylinder compression ignition machine, and, in cases where rotational swirl was used, anemometers were fitted in the cylinder while passing air through the valves, at the velocity one knew it passing under actual working conditions.

M. de Ganahl raised the point of what would be the effect of altering the conditions of test on the relationship of various fuel observations. He had tried altering the test conditions over a fairly wide range, *i.e.*, by completely cutting out heat and by putting in what might be called a normal amount of heat also by varying the ignition points round the cylinder. He had not found any measurable relative change at all; one could alter the conditions so as to shift the H.U.C. half a ratio up, but relatively they would still be within one-tenth of a ratio, so that he did not think one was likely to go astray appreciably there.

Mr. Moore asked the very difficult question of what would be the H.U.C. of a single valve and a distributing valve in series. He was afraid he had not tried that at all. It would depend a great deal on the position of the sparking plug, and of course on many other factors, and also on the amount of exhaust gas reintroduced into the cylinder from the capacity between the working valve and the distributing valve. He would expect a very high compression ratio to be obtained.

Mr. Moore, interposing, said 8 to 1 had been obtained.

Mr. Ricardo thought it would probably be a very good thing from the point of view of pinking. He had not made any observations on the Junker type at all, but that again ought to be worked on. It was, generally speaking, exceedingly difficult to get any observation on a 2-cycle engine, because of the varying quantity and temperature of the exhaust products left behind.

Mr. Beale asked a question which he should like to think over. It was a very interesting point indeed, but he did not feel competent to answer it at the moment. It opened up a very interesting line of thought which he should like to follow.

Mr. Rabson raised the question of the position of the spark. It made very little difference. In all cases the spark was approxi-

mately flush with the surface; in no case was it protruding far into the cylinder, nor was it ever pocketed right back. There might have been a variation of a sixteenth of an inch between the various engines in test. Certainly the mixture of air to fuel was constant in all cases, and the mixture he was quoting was one giving maximum power, which was approximately 15 per cent. excess of fuel. That was the mixture which gave the greatest tendency to pink. As a matter of fact, between a perfectly chemically correct mixture and one with as much as 40 per cent. excess fuel there was very little difference in the tendency to pink. Beyond the range the compression ratio went up a little with weaker or stronger mixtures, but one was safe in taking observation at round about the maximum torque mixture strength.

On the motion of the Chairman, a hearty vote of thanks was accorded to the author, and the proceedings then terminated.

The following written contribution was subsequently received from **Mr. W. Cochrane**:—

I read with interest Mr. Ricardo's paper on the above subject. I do not agree with him that detonation is the correct word to describe what takes place in the combustion chamber of an I.C. motor due to excessive temperature. Pre-ignition is the correct word to express what actually takes place. It is caused by the red hot exhaust valve functioning like the old hot tube ignition, igniting the charge before the piston reaches top dead centre, thus causing the motor to knock violently. At what point in the compression stroke pre-ignition begins no one knows, but it is reasonable to suppose that as the pressure in the cylinders gradually rises, so pre-ignition gradually spreads, and before the piston reaches top dead centre the charge is fully ignited, exerting its force in the wrong direction. No cylinder could stand the force of detonation or spontaneous combustion. The present system of poppet valve gear is unscientific; it separates the petrol from the air, and cools both instead of heating them when entering the cylinders. It expels the exhaust through a red hot valve and port which limits the compression ratio, causes pre-ignition, and the necessity to use a rich mixture, which in turn causes carbon monoxide gas in the exhaust. Hot spot induction and other theories advanced by Mr. Ricardo will never solve the problem of perfect gas distribution in poppet valve motors, nor will the various shapes of inlet manifolds, because the vapour has condensed back again into liquid petrol when it reaches the cylinders in unequal quantities, according

to the varying shapes of the inlet manifolds. This causes roughness in running, especially in six-cylinder motors. It is wrong to place the carburettor below the level of the valve ports, because petrol, being heavier than air, it follows that when drawn upwards it takes the shortest route, which is along the bottom of the pipe, thereby quickly separating itself from the air after leaving the carburettor as a vapour perfectly mixed with air. The successful solution of the difficult problems of perfect vapourisation, and equal distribution of the gas supplied, especially to six-cylinder motors, can only be accomplished by taking the gas downwards to the valve ports from an expansion box to which the carburettor is directly fixed. This means converting the dangerous carbon monoxide gas now contained in the exhaust into comparatively harmless carbonic acid gas. In the December issue of the *Automobile Engineer*, Mr. Whatmaugh, an eminent authority on pre-ignition carburettors and fuels for I.C. motors, describes and illustrates a high compression motor fitted with a new type of valve gear which solves the problems of pre-ignition, carbon monoxide gas in the exhaust, and the equal distribution and vapourisation of fuel, especially of the heavy type. I was interested in the new Ricardo head for poppet valve motors. It appears to me to be similar to the Hotchkiss head which has been on the market for several years. Perhaps Mr. Ricardo will explain the difference between the two heads, and at the same time correct any of the above criticism of the poppet valve motor which he may consider wrong.

The Author in reply wrote:—Mr. Cochrane objects to the use of the term "detonation," and goes on to say that what is commonly described as detonation is really preignition. Here he is very wide of the mark; detonation or "pinking" is entirely distinct from preignition; the former takes place solely at the latter end of the normal combustion process and has nothing premature about it, while preignition means ignition from some hot point or surface before the passage of the spark. It is perfectly easy to observe both phenomena and, in the case of preignition at any rate, to note exactly at what point in the compression stroke it starts. Preignition does not cause a ringing knock like detonation or pinking. It can be felt rather than heard, and is audible only as a faint dull thud. It is very seldom indeed that preignition occurs from a hot exhaust valve—in 99 cases out of a 100 the sparking plug points are the cause. The writer has run many tests with the exhaust valves water-cooled and has found practically no gain so far as preignition is concerned, and only a rather disappointingly small gain so far as detonation or pinking is concerned; thus, as between red-hot exhaust valves and stone cold in the same combustion chamber

the difference in H.U.C.R. is only about half a ratio. The writer holds no brief for exhaust valves—they are vile things, but don't let us exaggerate their villainy.

He fears that Mr. Cochrane is rather an optimist if he hopes seriously to improve distribution by allowing the mixture to flow downwards to the cylinders—gravity plays little part in the composition of a frog. Finally, Mr. Cochrane refers to the "new" Ricardo head and asks how it differs from that used in the Hotchkiss cars—it does not differ—this head, as mentioned in the paper, is now some ten years old and the Hotchkiss Co. are one of those, and by no means the first, who have employed it.

INSTITUTION OF PETROLEUM TECHNOLOGISTS.

THE ONE HUNDRED AND SIXTH GENERAL MEETING of the Institution of Petroleum Technologists was held at the house of the Royal Society of Arts, John Street, Adelphi, London, W.C. 2, on Tuesday, January 10th, 1928, Mr. E. H. CUNNINGHAM CRAIG occupying the Chair.

The Secretary read the following list of new members elected :—

The following gentlemen have been elected as :—

Members.—Albert Edward Atkinson, James Alexander Jameson, Edward Herbert Keeling.

Transference to Member.—Walter Edgar Aylwin.

Associate Members.—David Walker Alexander, Thomas Douglas, Walter Leslie Forster, Henry Ladislaus Jodkiewicz, John Trevarther Knight, Reginald Patrick Linstead, Charles Frederick Lloyd, Thomas Frederick Mason, Edward Ronald Plewman, Walter Claude Sydenham, Leslie Alfred Werrett.

Transference to Associate Members.—Geoffrey David Jardine, Adolf Rauch.

Students.—Arthur Patrick Chamberlain, John James Gladwell Clark, Charles Edward Hobley, Lawrence Fielder Stockdale, Hendrik Adriaan van Westen, John Granville Withers.

Associates.—Arthur Eaton, John C. Fell, Jack Cooper FitzHenry, James Oliver Innes, Charles Egbert Reynolds Sams, Donald James Walker, Arthur Charles Wilkes.

The Chairman said there was no need for him to introduce the reader of the paper, Dr. Dudley Stamp, who had already contributed a most interesting paper to the Institution on the subject of Burma. Dr. Stamp was a geologist who was not content with studying the mere superficial details of structures and of oil seepages, but who had boldly tackled the larger problems in connection with oilfields.

The following paper was then read :—

The Connection between Commercial Oil Deposits and Major Structural Features with Special Reference to Asiatic Fields.

By L. Dudley Stamp, B.A., D.Sc., A.K.C., F.G.S. (Associate Member).

I.—Statement of the Problem.

As long ago as 1855 the geologist, Godwin-Austen, predicted that a concealed coalfield would be found beneath the younger rocks in south-eastern England.[1] His prophecy was based on a careful study of the exposed coalfields of Wales and the west of England, on the one hand, and the coalfields of northern France, Belgium and Germany on the other. He noted the relationships between the then known coalfields and the major geological structure of the whole tract from Wales to Germany. It is unnecessary to detail the circumstances of the discovery of the East Kent coalfield, which so amply fulfils Godwin-Austen's prophecy. The main point of importance relevant to the present discussion is that there is no surface indication whatsoever of the hidden coal basin.

Is it possible that hidden oilfields exist of which there is, in the same way, no surface indication whatsoever? There can be little doubt that numerous such fields exist. Indeed, "wild-catting" has already succeeded in locating some. It is worth noticing, in the first place, that the majority of fields have been discovered because they are imperfect. Either the oil-retaining strata are sufficiently far from the ideal to allow a certain quantity of oil to escape to the surface to form an oil-seep, or part of the oil-bearing anticline or other structure has been worn away and exposed the structure of the ground and the nature of the beds. A really perfect oilfield, from which no trace of the oil had seeped to the surface, and no part of the structure had been denuded so as to demonstrate the possibly petroliferous character of the rocks, would be extremely difficult to detect. In many parts of the world there are vast stretches of alluvium, in other parts huge areas of blown sand or loess, which completely hide the underlying structure. In other areas, as will be mentioned shortly, comparatively simple folds are hidden by overthrust or over-folded masses of complex structure. It is in such areas that hidden oilfields may well exist; there are certain areas where they virtually must exist.

[1] *Quart. Jour. Geol. Soc.*, Vol. XI., pp. 533-6; Vol. XII., pp. 38-73.

It is not the purpose of this paper to attempt to predict where hidden fields may lie. Its object is to examine what is known of the relationships between existing oilfields and main structural features, and whether there is any hope that such relationships may be used to guide the search for hidden fields—or at least to place " wild-catting " on a scientific, geological basis.

II.—The Disposition of the World's Oilfields.

For the purposes of this study we may distinguish a broad, threefold division of the earth's surface into :—

(*a*) Plateaux of ancient rocks.
(*b*) and (*c*) Lowlands.
(*d*) Mountain chains.

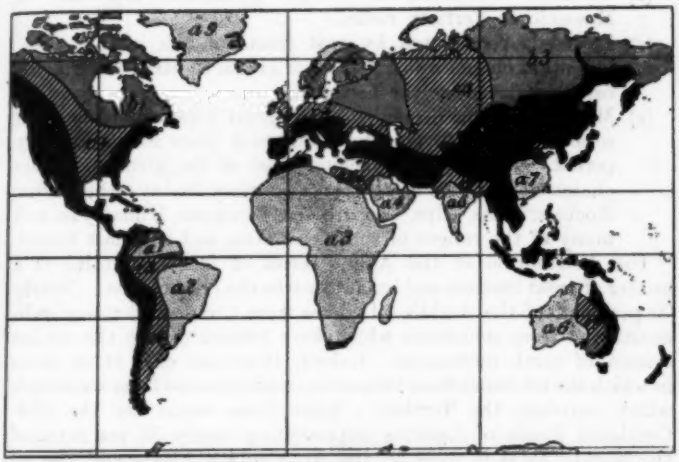

Fig. 1.

The main morphological units of the earth's surface :—
(*a*) Plateaux consisting mainly of ancient metamorphic rocks for the most part incapable of being petroliferous :
a^1 Guiana Highlands ; a^2 Brazilian Plateau ; a^3 African Plateau ; a^4 Arabian Plateau ; a^5 Indian Plateau ; a^6 Western Australian Plateau ; a^7 South China ; a^8 Scandinavian Highlands ; a^9 Greenland.
(*b*) Lowlands of ancient rocks, also non-petroliferous :
b^1 Laurentian Shield ; b^2 Baltic Shield and Russian Platform ; b^3 Angaraland :
(*c*) Lowlands of younger rocks which may be petroliferous :
c^1 Central Plains of North America ; c^2 Orinoco Basin ; c^3 Amazon Basin ; c^4 Argentine ; c^5 North European Plain ; c^6 West Siberian Lowland ; c^7 Plain of Hindustan ; c^8 North China Plain ; c^9 Australian Lowlands ; c^{10} Egypt-Iraq.
(*d*) Mountainous regions (in black). Oilfields, as shown in Fig. 3, tend to be concentrated on the flanks of such areas.

This broad division has been made in Fig. 1. Those areas constituting the ancient plateaux may, broadly speaking, be excluded in a consideration of the petroliferous regions of the world. It may be, of course, that small basins containing oilshales, or even superficial deposits with small quantities of free oil, may lie on their surface, but they are not the home of any major field.

Geologically the remaining areas, the lowlands and great mountain chains, may be separated into :—

(*a*) Lowlands consisting of areas of ancient rocks which, in the course of long ages, have been worn down almost to sea-level. The Laurentian Shield of Canada, the Russian Platform (constituting the greater part of Russia), and the ancient massif of Angaraland are outstanding examples.

(*b*) Lowlands consisting of younger sediments, especially of Mesozoic or Tertiary rocks.

(*c*) Lowlands constituted by vast tracts of alluvium.

(*d*) Mountain chains which owe their origin to earth movements earlier in date than the Tertiary.

(*e*) Mountain chains belonging to the great Alpine or Himalayan series of earth movements, which took place in the Tertiary period. To this group belong most of the great mountain chains conspicuous on the earth's surface to-day—the Andes, Rockies, Atlas, Alps, Carpathians, Caucasus, Himalayas, and many of the ranges of south-east Asia and the East Indies.

The distribution of the Alpine series of fold mountains is a matter of great interest and importance to the oil geologist. Nearly 70 per cent. of the world's oil comes from Cretaceo-Tertiary sediments and from structures which were formed during the Alpine system of earth movement. Indeed, there are only three areas in which the oil comes from Palæozoic sediments and from structures which antedate the Tertiary. These three areas are the Mid-Continent Fields of America (representing nearly 30 per cent. of the world's total in 1924-5), the Appalachian Fields and the oil fields of Ontario. Were it not for the Mid-Continent and Appalachian fields, one could truthfully say that all the great oilfields of the world are associated with the Alpine Mountain Chains.

Not only is there thus a very marked association of great oilfields with the principal Tertiary fold ranges, but a closer examination reveals also certain definite relationships between the oilfields and the main lines of folding within the folded belts. The fields invariably lie on the flanks of the folded belts. One obvious explanation of this phenomenon is that, where the rocks are intensely folded in the midst of the main ranges, the oil has literally been squeezed out. But, as will be pointed out later, there are other causes controlling the location of fields.

It has often been pointed out that there are frequently active or recent extinct volcanoes in oilfield regions. One's mind immediately flies to Burma, Sumatra, Java and Japan as examples. A map of the world showing the position of the principal volcanic areas seems to bear out the close association of vulcanicity and oil (Figs. 2 and 3). It is upon this association that theorists have averred that vulcanicity has played some part in the generation of oil—not necessarily that oil is of volcanic origin, but that its formation is due to some degree to the action of the earth's internal heat. When, however, one studies oilfield countries in detail,

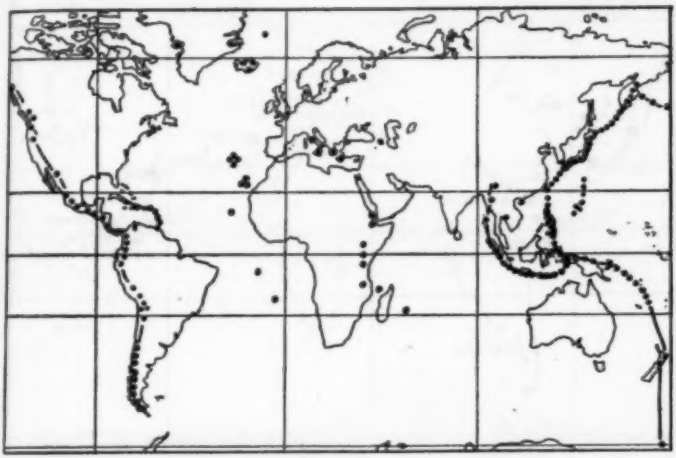

FIG. 2.

Map of the world showing the disposition of active and recently extinct volcanoes (generalised). It should be noted that the majority of volcanoes are associated with the mountain belts shown in Fig. 1.

one finds that the oilfields are rarely near the volcanic regions; strata elsewhere petroliferous become barren in the neighbourhood of volcanoes. This will be well seen later in the cases of Burma and Java. Evidence is not lacking that the association of oil and vulcanicity is accidental. Looked at in another way, oilfields are associated with the great belts of Tertiary folding; volcanoes are also associated with the same belts of folding. But there is no genetic connection between oil and vulcanicity—excluding local cases where the ignition of oil may have helped volcanic action, or where the heat from volcanoes has distilled small quantities

of oil from coal or shale—but rather an antagonism. Where one is found, the other is absent.

III.—The Origin of the Great Folded Belts.

Since there is definitely a connection between the great folded belts and the distribution of oilfields, it is of importance to consider the formation and structure of such belts. Whatever the type of fold belt, accumulations of oil of commercial importance tend to be found only on the more gently folded areas on the flanks of the belt.

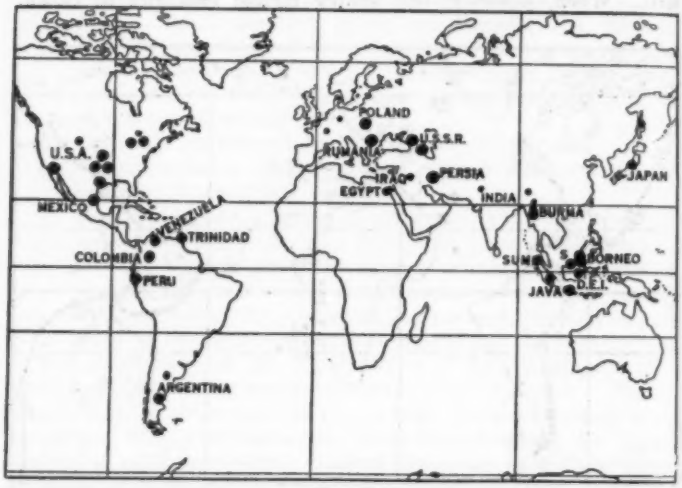

Fig. 3.

Map of the world showing the disposition of the principal oilfield regions. It should be noted that the majority of oilfields are associated with the fringes of the mountain belts shown in Fig. 1, and that many fields are closely associated with the volcanic lines shown in Fig. 2. (Figs. 1 to 3 are adapted from Stamp's "Commercial Geography," Longmans, Green & Co., Ltd.)

It is possible to consider the earth's surface as consisting of relatively stable blocks separated by lines or, more correctly, zones of weakness. We can follow Wegener and consider the stable continental blocks capable of drifting; we can follow the theories of Wegener as modified by Argand and conceive of stable continental masses separated by broad zones in which folding and other movements are concentrated; or we can uphold the old "orthodox" school of geology. But whichever view we take, we do not get away from the undoubted fact that it is the great oceanic troughs

of late Mesozoic and Tertiary times which have become the great mountain chains of to-day. Expressed in another way, it is the great geosynclinal areas which are the zones of weakness; the weakness increases in intensity from the margins towards the centres of the troughs, and it is the thick accumulation of marine sediments in the centres which eventually form the great mountain

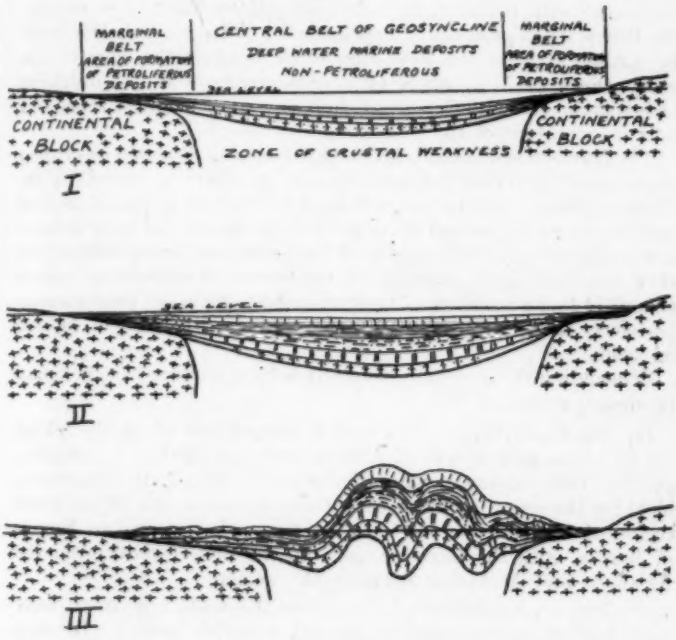

FIG. 4.

Diagrams illustrating the deposition of sediment in geosynclines and the formation of geosynclinal mountain ranges. Diagram III. shows the reason for the usual occurrence of oilfields in the minor folds on the flanks of the main folded region.

chains. The sediments laid down in the shallow seas, bays, gulfs and lagoons which lie on the margins of the old oceans are relatively less disturbed by folding. Three stages in the history of a typical geosyncline are shown in Fig. 4. The conception of the geosyncline is due to the great American geologist, James Hall, who in 1859 thus interpreted the Appalachian mountain chain. It should be

noted that the stable blocks on either side of the geosyncline need not, necessarily, be dry land. The succession of curved geosynclinal areas which flank the continent of Asia on the east may be conceived as lying between the continental mass of Asia, on the one hand, and a stable block forming the floor of the Pacific Ocean on the other.

There is strong evidence that the accumulation of oil is definitely associated with the marginal seas and gulfs of the former oceans. As Beeby Thompson aptly expresses it: "In general, the most favourable area for the formation of oil is along the edge of the geosynclines, . . . where oil is obtained from almost unfolded beds [as in the Mid-Continent Fields], . . . they represent lateral extensions of the geosynclinal sediments."[1]

The distribution of oilfields is thus determined by the actual geography of the period concerned, particularly, that is, with the geography of the Tertiary period. In the future it must be the task of the geological staff to use every method of research at the disposal of their science in reconstructing the geography of the geological period with which they are concerned. Change of conditions of deposition means change of facies; change of facies means migration of petroliferous horizons. In this direction lies the hope of discovery of hidden oil fields.

Considering the structure of folded belts a number of types may be distinguished:—

(1) The fan-structure, in which a central belt of highly folded strata, often with a core of ancient rocks, is flanked by smaller, gentler folds dying away *diminuendo*. Anticlinal structures, ideal for the retention of oil, thus occur on either side of the main region of folding. We have, as an example, the Arakan Yomas of Burma, with the Burmese fields on the eastern flanks and the Assamese and Arakanese fields on the western.

(2) The asymmetric type, in which the folds appear to have been formed by compression against a stable massif. In such cases open folds, suitable for the retention of oil, occur on one flank of the foldbelt (A—A in Diagram II. of Fig. 5). On the other flank suitable folds may occur over small areas, in some cases obscured by slight overfolding or reversed faulting. Underlying the great alluvial plains of Northern India there appears to be an extension of the ancient massif of Peninsular India. Against this the Himalayas have been folded and so commence abruptly. There is no belt of gentle, open folds suitable for the retention of oil except in the Attock district of the Punjab, where

[1] "Oil Field Exploration and Development," New Ed., Vol. I. (1926).

the Khaur Field lies in a position analogous to A^1—A^1 in Diagram II. or Diagram III. of Fig. 5.

(3) The overfolded and overthrust type, of which the Alps and Carpathians form outstanding examples. This type is a develop-

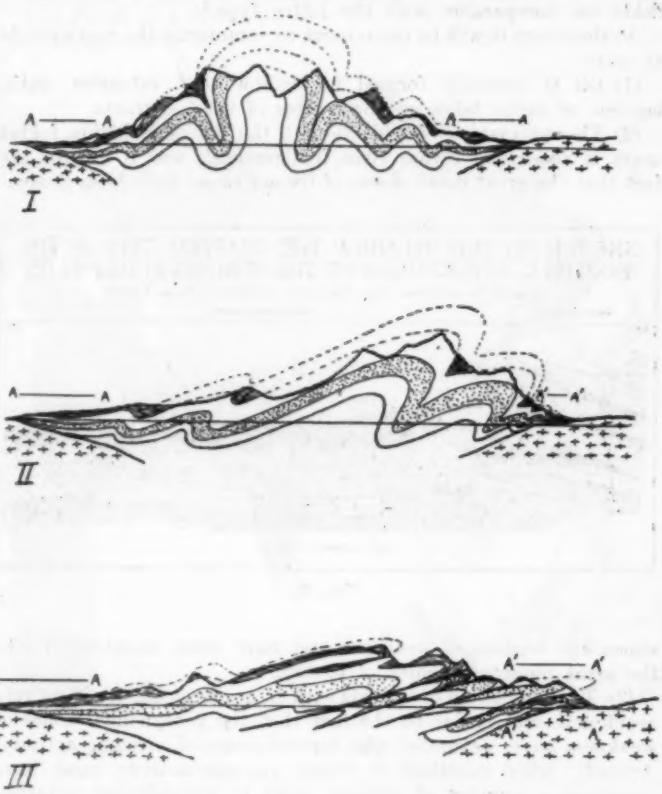

FIG. 5.

Three main types of folded mountain belts. The regions marked A—A are the areas in which oil fields are likely. In the cases marked A^1—A^1 the oilfields will be partly or entirely hidden by overfolded or overthrust older strata.

ment of the last and the Himalayan Chain might, perhaps more correctly, be included here. In many cases the folding is too intense to allow of the existence of oil fields; in other cases oil

may occur on the gently folded margin (A—A of Diagram III. of Fig. 5), or the gentle folds, suitable for the retention of oil, may be entirely hidden by overthrust masses (as at A^1—A^1 of Diagram III., Fig. 5). As shown in Fig. 6, the important Rumanian fields are comparable with the latter type.[1]

At this stage it will be convenient to summarize the main points at issue :—

(1) Oil is normally formed in shallow seas, estuaries, gulfs, lagoons or saline lakes on the fringes of oceanic tracts.

(2) The association of oilfields with the flanks of highly folded areas is accidental rather than fundamental, and is due to the fact that the great ocean deeps of former times have been geosyn-

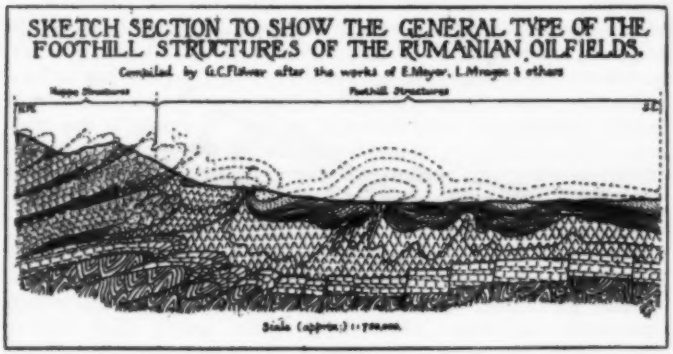
SKETCH SECTION TO SHOW THE GENERAL TYPE OF THE FOOTHILL STRUCTURES OF THE RUMANIAN OILFIELDS.
Compiled by G.C.Flower after the works of E.Mayer, L.Mrazec & others
Nappe Structures
Foothill Structures
NW.
SE.

Fig. 6.

clines and regions of weakness and have been transformed into the great mountain chains of to-day.

(3) The association of oilfields with areas of vulcanicity is entirely accidental, and is due to the fact that the geosynclinal zones of weakness have, naturally, also formed zones of volcanic activity. Actually, when examined in detail, volcanic activity must have destroyed a number of oilfields, since in petroliferous countries promising structures are barren when in the neighbourhood of volcanoes.

It is now proposed to put these assertions to the test by examining a number of the oilfield regions of Asia. Asia is chosen because it is the continent of which the writer has some personal knowledge ;

[1] "Flower : An Introduction to the Tectonics of the Rumanian Oilfields." *Jour. Inst. Petr. Tech.*, Vol. XI. (1925), pp. 61-75.

it will be left to others to accept or reject for other areas the premises here laid down.

IV.—Some Asiatic Oilfields.

(A) *Persia and Mesopotamia.*—The petroliferous belt of Persia and Mesopotamia affords an interesting example of position. To

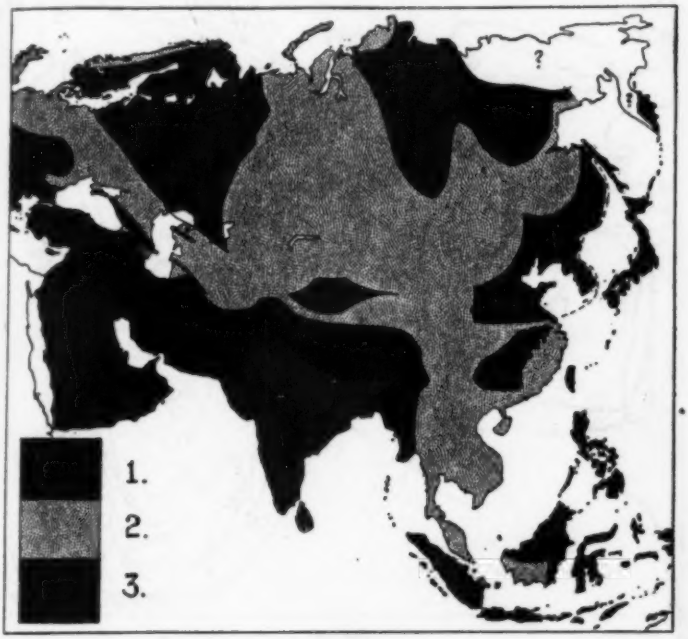

Fig. 7.

Structural map of Asia (based on Argand):
(1) Regions folded during the period of the Tertiary or Alpine movements, and consisting mainly of Mesozoic and Tertiary rocks originally deposited in geosynclines.
(2) Regions folded prior to the Alpine movements and consisting mainly of Palæozoic sediments.
(3) The ancient continental blocks, consisting mainly of pre-Cambrian metamorphic rocks.

the south lies the great plateau-massif of Arabia. This massif consists of a continental block of ancient crystalline rocks, thinly covered, especially towards the north, by Mesozoic and Tertiary

sediments. These latter must have been deposited on the edge of the continental mass, that is on an ancient continental shelf. By reason of their position, as resting on an ancient stable block, these later beds have been but slightly folded. It will be seen that they correspond in position with the slightly disturbed beds on the right of Diagram II. or III. in Fig. 5. It will be seen that

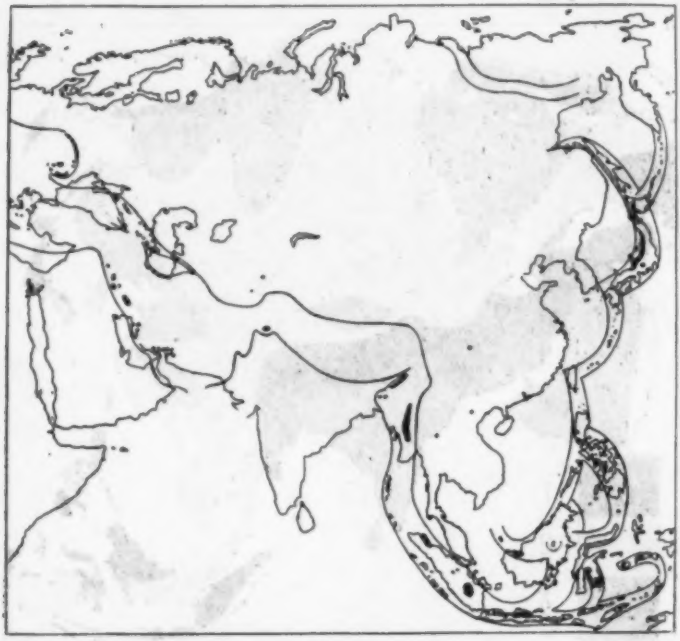

FIG. 8.

Map showing the disposition of oilfields in Asia. Oilfields shown in black. It should be noticed that nearly all the oilfields lie within the Cretaceo-Tertiary geosynclinal areas or marginal fold-belts, the boundaries of which are shown approximately by black lines. The principal oilfields lying outside these areas are the small fields of China.

they come within the possibly oil bearing region of that diagram, and thus it is found that certain of the oil shows of Mesopotamia—the more southerly ones—are found in these slightly disturbed beds which rest on the ancient rocks.

To the north of the ancient mass of Arabia, the fold mountains of the Alpine Belt of Persia commence with rather remarkable

suddenness. The fold belt extends right across Persia to the borders of the Caspian Sea and the lowlands of Turkestan. Broadly speaking, it seems permissible to liken the structure of the whole belt to Diagram II., or possibly III., of Fig. 5. The mountains of Kurdistan and Laristan are built up of highly folded Tertiary rocks;

FIG. 9.
Sketch map showing the relationships of the Persian and Mesopotamian oil belts.

to the north, occupying much of the central plateau, are huge stretches of rocks varying in age from Archæan to Jurassic (the 'Oman Series, etc.),[1] whilst still further north younger rocks again appear. The petroliferous belt is associated with the extensively

[1] See, *inter alia*, G. E. Pilgrim, *Mem. Geol. Surv. India*, Vol. XLVIII.; A. F. Stahl, Geologie von Persien, Handb. d. reg. Geologie.

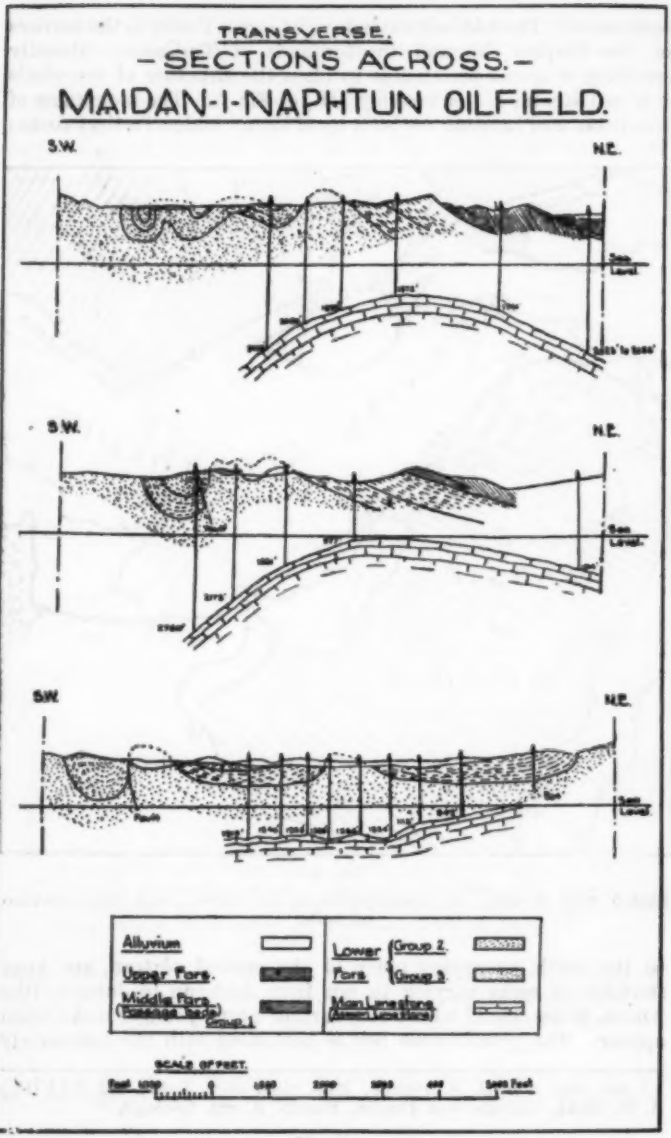

FIG. 10.
The structure of the Maidan-i-Naftun Oilfield, Persia (after Richardson).

folded Cretaceo-Tertiary belt, and thus corresponds with A^1—A^1 in Diagrams II. or III. of Fig. 5. There will be a difference of opinion whether one should liken the Persian belt to Diagram II.—where there is simple overfolding—or to Diagram III., where the overfolding is complicated by overthrusting. Of the overfolding there is no doubt (see Pilgrim, *op. cit.*, pl. 13). It is unnecessary to enlarge here upon the curious anomalies between the surface geology in the Maidan-i-Naftun field and the disposition of the main anticline in Asmari Limestone underground. Richardson's sections, here reproduced for easy reference, bring out the wide variations.

Three interpretations are possible :—

(*a*) That the anticline in the Asmari Limestone is a "buried hill."[2]

(*b*) That the softer beds overlying the Asmari Limestone have been independently and more extensively folded as a result of their lithogical character.

(*c*) That the folded series exposed at the surface has been thrust over an underlying group.

In the latter case the Persian field corresponds with A^1—A^1 in Diagram III. of Fig. 5. If extensive overthrusts do occur in Persia, a ready explanation is afforded of the fact that a number of surface anticlines have been drilled without discovering oil.

In passing, it may be noticed that the Oxus oil belt of Northern Persia and Afghanistan corresponds to the northern borders of the old geosyncline (A—A in Diagram III. of Fig. 5).

(B) *The North-West Indian Fields.*—In its broad general relationship, the petroliferous area of North-West India resembles that of Persia and Mesopotamia.

To the south lies the huge ancient continental block of Peninsula India, built up for the most part of pre-Cambrian crystalline rocks. This ancient mass extends northwards under the great alluvial plains of Northern India. It was against this stable block that the huge thicknesses of sediments deposited in the Cretaceo-Tertiary trough were crushed and folded to form the world's greatest mountain range, the Himalayas. In general the structure of the Himalayas is that shown in Diagram III. of Fig. 5. To the north of the Ganges Plain the folding is so severe that even the latest Tertiary rocks—the Siwaliks—are intensely folded and cut by thrust faults. Fig. 11, taken from the works of C. S. Middlemiss shows the extensive faulting even in the outermost zone. There is obviously but little hope of the preservation of oil in such highly disturbed

[2] S. Powers, "Reflected Buried Hills," *Bull. Amer. Assoc. Petr. Geol.*, Vol. X., p. 431.

strata. Similar intense folding and overthrusting occur in the older rocks of the Himalayas—as exemplified by the areas studied by Middlemiss and others, and of which an example, taken from the studies of Middlemiss, is shown in Fig. 12. Only in one region of Northern India is there an area where the folding is sufficiently gentle to allow of the existence of an oilfield of commercial import-

FIG. 11.

Section from the Middle Himalayas through the Outer Himalayas of Kumadu, showing the highly overthrust and overfolded nature of even the outer zones of the Himalayas. L.S., Lower Siwaliks; M.S., Middle Siwaliks; U.S., Upper Siwaliks. Thick black lines are thrust faults (after Middlemiss).

ance. That region lies in the North-West, in the Punjab, between the famous Salt Range and the Himalayas. The structure of that region corresponds very closely with A_1—A_1 in Diagram III. of Fig. 5. The area has been studied and described by Pascoe,[3] Pinfold[4] and more recently by Parsons.[5] Fig. 13, reproduced from one of Pascoe's sections, illustrates the general character of the

FIG. 12.

Section through the Central Himalayas of Spiti:—(1) Kioto Limestone (Jurassic); (2) Spiti Shales (Jurassic); (3) Giumal Sandstone (Cretaceous); (4) Cretaceous Flysch; (5) Igneous. T—T, Thrust Fault (after Von Krafft).

folding and thrust faulting in north-west India. It is interesting to note that Parsons believes the thrust faulting to be much more important and much more widespread than previous observers have

[3] *Mem. Geol. Surv. India*, Vol. XL.
[4] *Rec. Geol. Surv. India*, Vol. IXL. (1918); *Jour. Asiatic Soc. Bengal*, N. S., Vol. XIV. (1918).
[5] *Jour. Inst. Petr. Tech.*, Vol. XII., pp. 439-505.

FIG. 13.

Section through Chak Dalla (after Pascoe) to illustrate thrusting and overfolding in the north-west Indian oilfield region.

believed. Differences of opinion on this point do not affect the general relationships of the area to the Himalayan geosyncline and the ancient continental block of Peninsular India. The main outlines of the geotectonics have been given by Pascoe *op. cit. sup.*, pp. 445 *et seq.*). Pascoe considers that the Himalayan syncline had already "buckled" to such an extent that the tertiary deposits along the northern shores (Oxus Oil Belt) were never continuous with those along the southern shores (Punjab Oil Belt). In other words, the oil-bearing deposits were laid down in epicontinental seas or gulfs, not on the flanks of a geosynclinal ocean, but on the flanks of the region where the geosynclinal ocean had been. This is of considerable importance because it has been suggested by some that the presence of oil on the margins of geosynclines is due to the migration of the oil from the central parts of the troughs, where it might have been formed, towards the margins.[1] This is obviously impossible where the centre of the geosyncline had been folded *before* the depositing of the principal petroliferous strata on the margins.

(C) *Assam and Burma.*—If one were dishonest, one could point out how perfectly the arrangement of the oilfields of Assam and Burma accords with the general principles already enunciated. For the mountain wall which separates Burma from India, and of which the southern portion is known as the Arakan Yoma, may be regarded as a highly folded geosyncline. It has the typical core of ancient rocks and is flanked by folds of younger and younger rocks. The oilfields of Assam and the Arakan Coast lie on the western flanks, the oilfields of Burma proper on the eastern flanks. Further east lies the old stable continental block of the Shan Plateau, and its continuation into Tenasserim and Malaya. One could point out how the Burmese oil-bearing beds were formed along the eastern margins of the geosyncline and the Assamese beds along the western margins.

There can be little doubt that the Arakan Yoma range of mountains does represent a buckled geosyncline which lay between the Indo-Malayan (Shan States—Tenasserim—Malaya) continental block on the east and a stable massif on the west. The writer is inclined to regard the stable block on the west as being a foundered block now underlying the Bay of Bengal. The Jurassic sea spread over part, at least, of the Shan States, and it would seem that the geosynclinal trough was most clearly defined during the Cretaceous. At the end of the Cretaceous or in early Eocene times the trough had commenced to buckle, and the geosynclinal ocean was replaced

[1] Clark, "Organic Theories of Oil Origin," *Jour. Inst. Petr. Tech.*, Vol. XII., p. 275.

by two comparatively shallow gulfs; both closed by land to the north and open to the sea to the south. These two gulfs were the Assamese Gulf and the Burmese Gulf. Into both these emptied great rivers flowing from the north or north-east. Each gulf behaved as a small geosyncline; indeed, the Burmese gulf itself

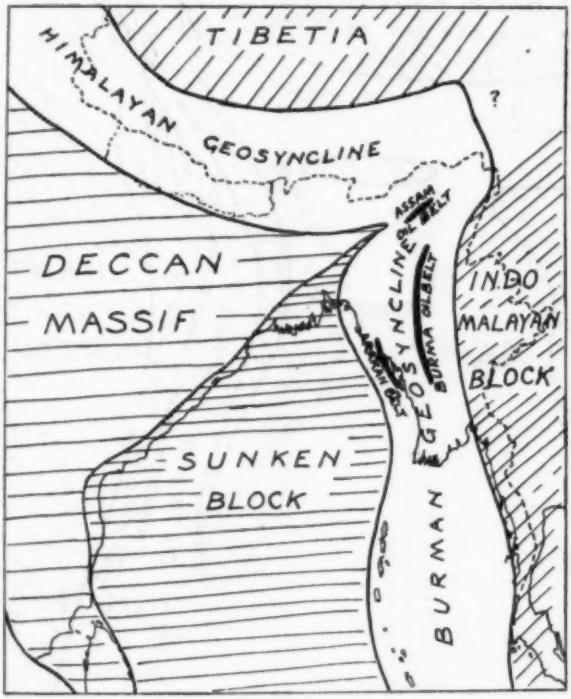

FIG. 14.

Sketch map, showing the relationships of the Assamese and Burmese oil belts

was almost certainly buckled at any early stage into a twin gulf with a ridge running down the centre separating it into an eastern and a western half.

In a former paper read before this Institution,[1] the writer attempted to show that the history of tertiary times in Burma

[1] "Conditions Governing the Occurrence of Oil in Burma," *Jour. Inst. Petr. Tech.*, Vol. XIII. (1927).

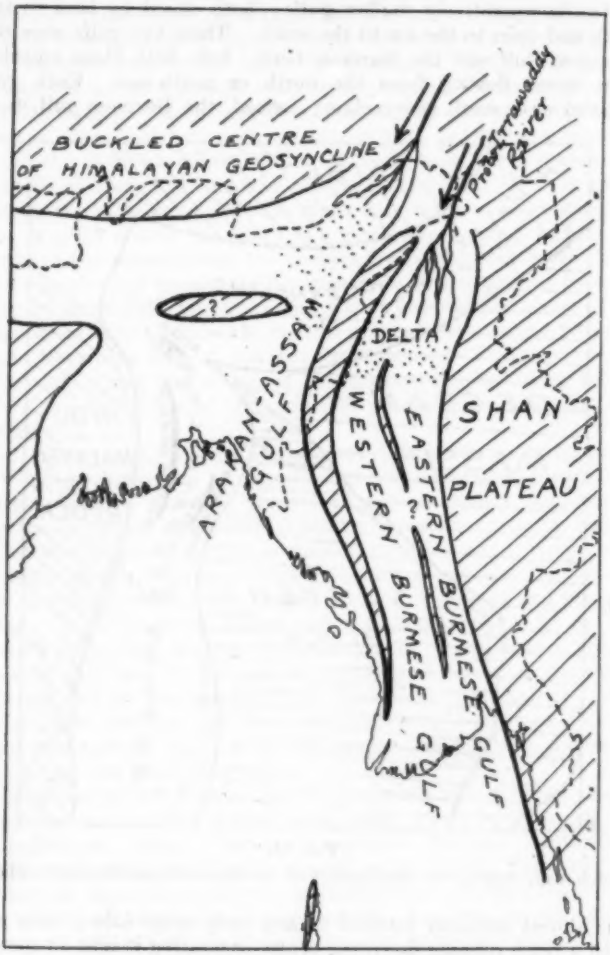

FIG. 15.

The early tertiary geography of Assam and Burma.

consisted of the gradual filling up of the Gulf, interrupted by intermittent backward migrations of the sea owing to the deepening of the trough. It was further shown that the deposition of oil

FIG. 16.

Section through the oil belts of Assam, Arakan and Burma:—(1) old crystalline rocks; (2) various pre-tertiary rocks; (3) Eocene; (4) Peguan (the principal oil bearing series); (5) Irrawaddian; black, volcanic. On the Arakan side only the general character of the folding has been indicated.

measures was definitely associated with shallow, brackish water conditions, that oil was absent from freshwater deposits and from the deeper water marine deposits.

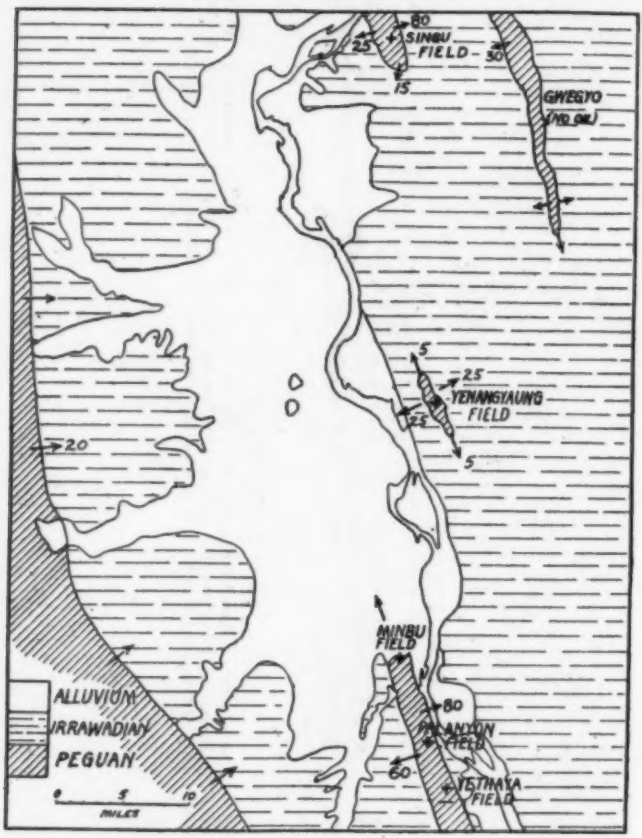

FIG. 17.

Sketch map showing wide gathering ground, Yenangyaung, and relation of neighbouring folds. Dips are generalised.

In Burma and Assam the oil measures can, it is true, be connected with the margins of an old Cretaceous geosyncline, but actually they were laid down in shallow, geosynclinal gulfs which arose

side by side as a result of the buckling of the major geosyncline. Figs. 14 and 15 illustrate these points. In the Burmese Gulf conditions suitable for the formation of oil existed roughly in the centre of the geosyncline, and it is there that the principal fields are found.

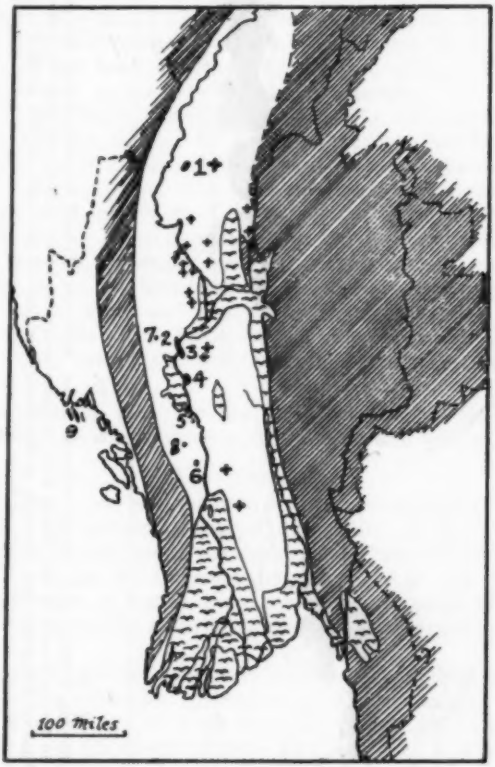

FIG. 18.
The volcanic regions and the oilfields of Burma.

The relation of the Assamese and Burmese fields to the main belts of folding is shown in section in Fig. 16. It need scarcely be pointed out that the oilfields are all on anticlinal structures, and that, other things being equal, the importance of the field

FIG. 19.

Map of Java showing Tertiary and recent volcanic rocks in black. It will be noticed that the petroliferous belt lies in the only part of the island from which volcanic rocks are absent and that the most important pools (those of Tinawoen and Panolan in Rembang, yielding 80 per cent. of the total, the position of which is indicated) are well removed from volcanic rocks. Other oil pools lie near Semarang; west, south-west and south of Surabaya and in Madura.

varies as the size of the gathering ground. Fig. 17[1] shows the wide gathering ground of Yenangyaung.

It will be noticed that in the north-east corner of Fig. 17 there is marked the Gwegyo inlier, in which no oil has been found. There are in Burma several other Peguan inliers which, though the lithology of the beds seems suitable, are barren of oil. It is found that closely associated with several such folds are the late tertiary or post-tertiary volcanoes of Burma. The writer has little doubt that the oil has been destroyed by volcanic action. Fig. 18 is almost sufficient by itself to prove this assertion.

(D) *The East Indies.*—Referring back to Fig. 8, it will be noticed that the various oilfields of the East Indies—Sumatra, Java, Borneo, etc., are shown as associated with narrow geosynclinal areas. These geosynclinal areas occur as a succession of festoons or arc round the south-east coast of Asia, and the writer is inclined to suggest that they are comparable with the Burmese area in that they lie between the stable continental mass of Asia, on the one hand, and stable but foundered blocks which underlie the oceans and seas to the south and east. It is further suggested that the East Indian fields will be found to lie in the midst of geosynclinal troughs closely comparable with the Burmese Gulf in magnitude and character.

A map of Java (Fig. 19) has been included as yet another demonstration of the separation of volcanic and oilfield regions.

V.—Summary.

1. An attempt has been made to analyse the connection between oilfields and geosynclinal areas of deposition. It is found that oilfields do frequently lie on the margins of old geosynclines, where the deposits in the centre are of deep water type. On the other hand oilfields, as in Burma, may occur in the middle of smaller geosynclines, where the latter are infilled by suitable shallow water deposits.

2. There is no genetic connection between oil and belts of folding (geosynclinal or otherwise) or between oil and vulcanicity.

3. The formation of oil is controlled by the geographical conditions during the period concerned. It is true that it is irrational to demand "any particular set of deposition conditions for petroleum origin," but certain essentials are required:—

(*a*) Abundance of organic material.

(*b*) Conditions suitable for rapid sedimentation and entombment of the organic material before its destruction by aerobic bacterial action.

[1] Reproduced from Stamp, "The Geology of the Oil Fields of Burma," *Bull. Amer. Assoc. Petr. Geol.*, Vol. XI. (1927), p. 570.

Suitable conditions are most typically found in gulfs, lagoons and epicontinental seas, which may or may not be connected with major geosynclines.

4. The great work for the geologist in search of hidden fields should be the reconstruction of the geography of the periods concerned. He will then be able to predict the lateral changes to be expected in the various beds when traced underground; he will be able to state in which direction they may be expected to become most richly petroliferous. The location of the actual structures may be assisted by careful observations of surface folds, but given that the geologist can state in which direction the possibilities of oil in quantity increase, geophysical methods should be able to assist greatly in locating the actual structures. To take a concrete example: where a possibly petroliferous series disappears under a tract of alluvium the geologist should be able, by a wide survey of surrounding tracts, to say in which direction chances are best, an underground anticline below the alluvium may possibly be located by geophysical methods. The two points to be emphasized here are:—

(*a*) The need for a wide geological survey, not restricted merely to beds believed to be petroliferous.

(*b*) The reconstruction of the former geography as a very essential part of the geologists' work.

VI.—Conclusion.

Perhaps some apology is needed for the present paper. It may be said, with some justice, that it contains little but a reiteration of matter which has long since entered the domain of the text-book, and which is common knowledge to any student of modern petroleum geology. Three reasons may be given for venturing to set out these broad generalisations before the Institution of Petroleum Technologists. In the first place, if a theory such as the association of oil fields with geosynclinal margins is in danger of becoming a text-book dogma, it is usually time it should be afforded a careful and critical examination. This paper will have served its purpose if it gives rise to a critical discussion on the subject. In the second place, it seems desirable that, if the theory can be really satisfactorily established, attention be focussed on its practical applications in the search for oil in still little known countries. In the third place, the attempt to apply the theory to Burma brings out the need of a modification which is probably of wide application, and which focuses attention on quite a different aspect—that of palæogeography.

DISCUSSION.

The Chairman said he was sure the members were deeply indebted to the author for a most interesting and intriguing paper. He thought there would be a very general agreement with most of the statements the author made, especially his conclusions. He had some comments to make on the paper, but he proposed to defer them to a later stage of the discussion.

Major T. R. H. Garrett, in opening the discussion, congratulated the author on a most interesting and useful paper, and one bound to cause a good deal of thought. But he could not agree with the author in some of the details. Fig. 6 is not typical of the known Rumanian Oilfields, and although this idea of the structure has been held by some, much money has been wasted in drilling holes in the ground based on this idea in the vain search for beds which never existed. He asked the author the grounds for showing in Fig. 7 south-west Borneo as consisting mainly of Palæozoic sediments, and assured him from personal observation that there is a large amount of folded coal-bearing beds of Eocene to Pleistocene age, and stated that perhaps more than one-half of this area is of Tertiary to Recent age. He pointed out that in Fig. 8 the Sarawak Oilfield is incorrectly shown as lying round Kuching, whereas it is over 300 miles to the north-east. He also asked why the large fields of Kutei are shown in this diagram as of less importance than those of Sarawak, Langat or Palembang. He asked whether the author was seriously of the opinion that the heat of Popa had raised the temperature at Gwegyo to a minimum of 350° C. required to distil the oil in the beds here. The distance is about 40,000 ft., and the intervening beds have a heat conductivity of only about one-twenty-fifth of that of copper. He suggested that the temperature of the beds at Gwegyo would not have been raised 10° C. by the volcanic activity at Popa. Further his recollection of a visit to Gwegyo in 1910 was that several wells had been drilled here, and that all had had shows of oil and gas, and that one had given a daily production of some tons, but that the quantity had not been considered commercial. His contention was that if the heat of Popa could have distilled any of the oil at Gwegyo it would have distilled all of it to say nothing of the gas. With regard to Java he had witnessed several eruptions of large volcanoes and would mention especially that of Tangkuban Prahu. Certainly where the hot material had come into actual contact with the quinine and lower down the tea growing on the slopes of the mountain some little damage was done, but none at all to the coffee, pepper, and rubber growing lower down, and the damage done was very small. If therefore the heat of a large volcano was insufficient to damage

the roots of the cultivation on its slopes surely it could not affect the oil in beds twenty miles away.

Sir Frederick W. Black, K.C.B. (Past-President), said the only remark he desired to make was to express his great pleasure that a paper had been read dealing with geology somewhat akin in nature to papers that he sometimes tried to prepare in relation to the economics of oil. Those interested in the commercial side of the question were in the habit of surveying the situation for varying periods. They looked for their probable supplies two, three, five, ten years ahead and sometimes for a longer period; and it was something of that nature on the geological side that had been presented to the Institution that evening. Broad features of the situation had been placed before the members in such a way that they were intriguingly near to subjects in which commercial men took an interest. The author made no attempt to show where oil would be looked for in the immediate future, but he did get away from particular fields and took a broad survey of the world from one end to the other, arranging the structures with reference to the general degree of their probable petroliferous content. In looking at the map of the world there were immense areas in regard to which people asked whether they were ever going to get below the alluvial deposits in tropical countries and find out whether oil existed or not. He congratulated the Institution on having had such a broad, general survey placed before it in relation to the geology of particular fields which brought them a little nearer to those considerations that business men had to keep in view when they were looking for fresh fields of supply.

Mr. T. Dewhurst said he was inclined to agree with the author's view that the paper was somewhat text-booky; in fact, he might even say that much of the subject-matter of the paper did not fall strictly within the realm of petroleum technology, but dealt rather with the general geological background or setting of the subject. He agreed with the author that there was no direct inter-relation of cause and effect between oil belts and volcanic belts. Volcanoes and oil deposits were, he thought, independent effects of various causes, and he was inclined to regard them as being even less related to each other than did the author. In that connection he desired to criticise the author's general conclusion that volcanic activity had undoubtedly destroyed a number of oilfields, and thought it would be necessary for the author to offer far more evidence than was in the paper before he could claim that the conclusion was in any way established. Personally, he did not accept it as a general proposition nor did he consider it to be true in the special case of Burma, either in the Lower Chindwin

or at Gwegyo. He agreed with Major Garrett in being unable to accept the view that oil had been driven from Gwegyo by the heat from Popa. The main problem of the paper was that of the occurrence of oil belts along the margins of geosynclines. They often heard of major geosynclines and major geanticlines, but very rarely of the intermediate and particularly the early stages in the evolution of the latter from the former, and although the subject was very speculative it was so very important that he ventured to make a suggestion regarding it. He believed that at a very early stage in this evolutionary process the central portion of a major geosyncline yielded to earth movement, so that the floor became uneven and corrugated, breaking up the geosynclinal surface into numerous small secondary geosynclines, basins and other depressions. Further movement would buckle up the middle portion of the major geosyncline so that it was only the secondary depressions situated on the more stable geosynclinal margins which had an opportunity of being relatively permanent and, therefore, of becoming the sites of oil-forming activity. He believed that the oil was formed in the centres of such secondary depressions and migrated towards the margins. In regard to palæography, he understood that most geologists had long ago accepted this as a convenient peg on which to hang innumerable details and which provided a geologist with a general picture of the conditions of deposition in any area which he happened to be studying. The references to palæography were, therefore, not new, but were nevertheless useful. There were several other points in the paper, notably the question of the structure of the Persian fields, which however important in themselves were relatively small questions compared with the main subject of the paper. In conclusion, he desired to say that, apart from the author's statement that volcanic activity had been responsible for the destruction of many oilfields, he was in complete agreement with the three propositions laid down on page 36, and which constituted the main conclusions of the paper.

Dr. J. A. L. Henderson thought that it was always useful to periodically collect for review theories which had been more or less generally accepted and standardised through inclusion in text-books and by publication elsewhere. During the course of one's work a continuous readjustment of viewpoint and ideas took place, particularly in regard to geological considerations. So far as the author's general thesis was concerned, he gathered that the author had tried to establish as a fact that most of the petroliferous deposits were formed only at the margins of geosynclines, and that the rocks within the basins would not

be oil-bearing. With regard, however, to the restriction of the zone of deposition or accumulation of petroliferous deposits to the margins of geosynclines, he could not see that any permanent marginal zone could exist, as during the course of the filling of a geosyncline by sediments constantly moving marginal zones towards the centre would result. This is illustrated by the occurrence of the petroleum deposits within the great Appalachian syncline, for instance. He did not think, therefore, that the picture presented in Fig. 4 adequately represented what the author probably intended to mean, as one would expect to find oil deposits or evidence of such in the highly flexured central regions depicted, as well as at the original margins. He quite agreed with the author in his concluding remarks that it was necessary to take the whole subject and to view it from a practical standpoint.

Mr. G. M. Lees said there were many points of detail which might be raised, but in the first place he desired to take exception to two of the author's major conceptions, first and foremost his conception of a geosyncline, which was shown diagrammatically in Fig. 4. Of course, the author was trying to generalise, and that figure was a generalisation, but as such it should coincide with facts as far as possible. Actually he would say it did not coincide with facts. The geosynclinal areas were not oceans and never had been oceans, and they did not bear any relation whatever to the present-day oceans. The Mesozoic geosynclinal sea was in the nature of the present-day Mediterranean; it had various islands scattered throughout its central regions, and nowhere any great depth. There are certain deposits often regarded as indicating abyssal conditions, the so-called radiolarites or radiolarian cherts, but the manner in which these rocks are usually interbedded with sandstones and brecciated limestones contradicts that view. The conception of a geosyncline as a gradually sinking area in the centre of which great thicknesses of sediments were formed was misleading. Taking the alpine geosyncline, in certain zones of the Alps, the Dachstein zone, for instance, there was a total deposition of 6,000 feet of limestone. That was the total sum of the sedimentation in that zone before the first intensive orogenetic movements took place. In another zone, the Hallstadt, the total sedimentation did not amount to more than 2,000 ft., so that if one were to draw such a diagram and show 2,000 ft. thickness in relation to breadth, it would have not the thickness of the line indicating sea level. The sedimentation of the geosyncline was not the fundamental factor, but rather

something more deep-seated. It was quite immaterial whether or not there was a thick deposition of sediments. The greatest deposition in all cases took place towards the margin, which was due to the greater amount of sand brought in from either side. In his opinion the author's diagram 3 in Fig. 4 was absurd. Although it was diagrammatic, such a state of affairs as that depicted had never taken place. There was no mountain range which had a structure anything like it. In all the mountain ranges of the Alpine system, without exception, folding had taken place in certain definite phases. For instance, there was a very definite phase which took place between the lower and the upper Cretaceous, the pre-Gosau movement. That phase could be traced right throughout the Alps. In the Swiss Alps proof of it is absent, though it doubtless had occurred, owing to the absence of unconformable upper Cretaceous rocks; but in the East Alps, through the Carpathians, through Persia and Afghanistan into the Himalayas and Burma, the East Indies and through to New Zealand, the Cretaceous phase was of great importance. The intensity of this phase was often such that great overthrust sheets or nappes were formed. Later sedimentation and movement was often limited to narrow zones within the greater system. A typical mountain system was double-sided, not as shown in diagram 3 of Fig. 5, and he would be very much surprised if the author could point out a single case in the whole Alpine system which corresponded to that figure. The Himalayas, for instance, were thrust strongly towards the south along the southern margin, but also along the northern margin of the Tamirs there was as great a thrust in the other direction, namely, towards the north. In a lecture recently given by the Mushketov Director of the Russian Geological Survey he described very vividly the extent of those northerly movements. Although he knew that the author's diagrams were merely diagrammatic, he thought a little greater attempt should have been made to make them coincide with geological facts. The points he was raising were of a geological nature and did not really concern the author's contention that the oilfields were going to be found in the outside zones. That happened for a variety of reasons, principally tectonic, but there was no fundamental reason why oil should not have been formed in the centre of the geosyncline. For instance, in the Tyrol there were very rich oil-shales which occurred right in the centre of the alpine geosyncline. The fact that they occurred in a shale-limestone series and not in a sandy facies governed the fact that they could never yield oil. The remarks the author had made in regard to palæography were very important, but he

thought most geologists already took such matters into consideration. One point the author did not mention was the importance of climate. Throughout geological ages the climate seemed to have been comparatively uniform throughout the world during the Palæozoic and the greater part of the Mesozoic eras. If one undertook a search for Palæozoic oilfields, one is not debarred from searching for them at the South Pole. But as time went on climatic zones became more differentiated. In the upper Cretaceous, and still more so in late Tertiary time, there were definite climatic zones. In the Miocene and Pliocene periods they were very well marked, so that no geologist need go to the South Pole to search for oil of that age.

Mr. J. Romanes said, although Dr. Stamp points out that in certain cases the main geosyncline may have buckled and formed a land mass before the marginal petroliferous facies set in, this does not seem to me to affect the main principle but to be merely a local feature. He need not accuse himself of any dishonesty if he did claim the Assam and Burma fields as a very beautiful example of marginal oilfields on either side of a main geosyncline. It is very interesting to note that Dr. Stamp claims that there is no genetic connection between oil formation and folding. The opinion has often been expressed that the actual dynamic forces involved in the folding movement were a factor in oil formation either directly owing to pressures set up or indirectly due to heat evolved, but he has certainly strong evidence in his favour in the little folded Mid-Continental fields of America.

There is another subject which Dr. Stamp merely touches on incidentally as it does not come strictly within the scope of the paper, but which is of importance in considering the whole question—namely, the distribution of oil-shales.

If the occurrence of oil-shales is viewed from the same tectonic standpoint, a striking difference is at once noticed. In the first place petroleum is essentially of marine or at least brackish water origin, whereas, although marine oil-shales undoubtedly occur, many are certainly of fresh-water origin.

A study of the distribution of oil-shales does not show the marked parallelism with the geosynclines, as in the case of the oilfields; on the contrary, there is a distinct tendency for them to belong to the continental areas, as, for example, their frequent occurrence among the "continental" deposits of "Gondwana-land." This first appears to negative absolutely theories which have been put forward ascribing the origin of oil-shale to the inspissation of an oil-soaked shale, and is incidentally a strong

argument against the practice of certain American writers of classifying oil-soaked shales with true oil-shales.

In conclusion, he thought that Dr. Stamp had opened up a line of investigation which was likely to prove most valuable both scientifically and economically.

The Chairman (Mr. E. H. Cunningham Craig) said the author had taken a well-known thesis and had illustrated it most excellently from countries, most of which he knew well. Papers of the kind that had been read that evening enabled the members to clarify their ideas considerably on the subject. Until Mr. Lees spoke there seemed to be a general misunderstanding as to what a geosyncline was. A geosyncline was an area of active movement downwards. Beside it was an area which was rising, and the centre of the geosyncline was where the thickest mass of deposit was formed and not in the centre of the sea or ocean between continental masses. Later on in that thick deposit, due to causes as to which authorities differed, the great start of the geanticlinal movement took place. He agreed entirely with what Mr. Lees had said in regard to some of the sections shown in Fig. 4. He deprecated to some extent the use of the terms "continental drift" and "zones of crustal weakness." He always thought that when such terms were used the users were plunging rather deeply into speculative theory. Of course, in their university days when they were slightly intoxicated by the teachings of Suess, Neumayr, Hein and the other great generalisers, the best and the least known of whom was De Beaumont, they dealt with continents and basins in a rather reckless manner. On examining countries, however, for general geological data which enabled oilfields to be discovered, it was necessary to get away from the higher and more wonderful theories and to place reliance only in the facts that could be hammered out. Three ages of mountain building movement were known. There was a fourth, but it was so old that very little was known about it. There were the mountain ranges that were formed in those days as geanticlines out of geosynclines, and there were oilfields in all those three. Those that were known most about were the latest, because they existed in a way in which they could be seen more easily. Those three movements existed and the oilfields were preserved on the flanks of them. That did not mean, however, that the oilfields did not exist at one time in the middle of them, but they were only found preserved on the flanks owing to elevation and denudation and also sometimes to considerable thrusting movements. That applied particularly to India, Java, Burma, Persia, the Andes and the Rocky Mountains, in fact to every country in which oilfields were known to exist. The only other point he

desired to mention was in connection with the eastern part of the Burmese basin. It was quite a shallow part of the basin; in fact it was probably land during a large part of Peguan times. He could assure the author that there was excellent evidence pointing to the fact that oil never existed in that part of the basin at all, indeed for some distance to the west of that basin there probably never was any oil in commercial quantities. He was sure it was the opinion of the members that the least they could do was to offer a most hearty vote of thanks to the author for the most interesting paper he had given.

Dr. Dudley Stamp, in reply, said that he would endeavour to deal with a few of the large number of points which had been raised in the course of the discussion. He was not responsible in any way for the section of Rumania to which Major Garrett objected; he simply borrowed it from the Journal of the Institution in order to illustrate one of his points. He might perhaps have made a happier choice by borrowing an illustration from another region. With regard to Major Garrett's remarks on South-west Borneo, he thought they were an excellent example of the advantage that would be gained if geologists who had practical knowledge of that country placed their views before the Institution. He had found it very difficult indeed to get anything beyond the most general statements with regard to the structure of Borneo. He had endeavoured to use such general statements as he could obtain, and they had been incorporated in his map; but if Major Garrett had actual facts which he could place before the Institution which would enable the members to improve their knowledge of Borneo he was sure they would all be grateful to him. It was important, of course, to distinguish between an old shield partly covered by later deposits and a folded zone. With regard to the small size given to the fields on the eastern side of Borneo, he referred Major Garrett to Fig. 3 of the paper rather than to Fig. 8. With regard to the disputed question of volcanoes having destroyed a number of fields, that was not in any case an essential point of the paper. There was one point, however, he desired to make in that connection. Both Major Garrett and Mr. Dewhurst had rather stressed the question of the distillation of oil by volcanic heat. Personally, he was always under the impression that oil was capable of burning, and he rather thought that the burning that took place in connection with volcanoes underground could not be judged by the action of a few ashes which were thrown out and happened to wither up a few leaves on a tea bush. He need scarcely say that he was in hearty agreement with Mr. Dewhurst on the point he had raised concerning the stages from the major geosyncline to the major

geanticline. Mr. Dewhurst had very ably expressed in a few words what he (the author) had rather haltingly tried to do in connection with Burma, pointing out that Burma did give an illustration of some of those stages; and that in the three gulfs side by side which he mentioned in connection with Burma and Assam there was an example of the stage between the major geosyncline and the geanticline. He thought, however, he must take exception to Mr. Dewhurst's remarks regarding palæography. In the course of his short geological career he had had quite a number of fierce quarrels with various people, one of the biggest quarrels being on the question of palæography. He had written a book on stratigraphy from the standpoint of palæography which had made a number of converts and a number of decided enemies. He thought he might be permitted to remind the members that very many geologists were brought up in schools in which they were taught not to venture into such wild realms of theory as palæography. They knew the geological sequences and described them, and that was sufficient. If he had stressed the question of palæography it was because he had suffered sometimes from the opposite point of view, which still regarded any attempt at the reconstruction of conditions as they were as being matters to be discussed behind closed doors rather than in the open. In reply to Dr. Henderson, who referred to the oilfields on the margins of geosynclines and queried whether there was such a thing as a marginal zone, he thought the best answer to the point was contained in the paper where he had stated that suitable conditions were most typically found in gulfs, lagoons and epicontinental seas, which might or might not be connected with major geosynclines. He allowed for the migration of the major geosyncline by emphasising that it was a question of geographical conditions and not the position of the centre of the geosyncline. He had quite enjoyed the attack that had been made on the paper by Mr. Lees. First, with regard to his conception of a geosyncline, he heartily agreed with Mr. Lees in the separation of what might be called the major oceans. He scarcely liked to call them the permanent oceans because that was begging the question. But there was a big difference between the major oceans that existed at the present day and what might be called the geosynclinal ocean. He thought the analogy Mr. Lees had drawn, that his (the author's) geosynclinal oceans were much more comparable with the Mediterranean Sea was quite justified. He followed Mr. Lees in that point, but perhaps he could not think of a word which meant something more than a small sea but something less than an ocean. If there was such a word he would like to use it in connection with a geosyncline rather than to misuse the word ocean. If he used the word sea he would be confusing the issue,

because there were many insignificant tracts of water which were dignified by that name. He did not follow Mr. Lees entirely on the point he made with regard to the thickness of sediments in geosynclines. Some, it was true, might have only 2,000 ft. of sediment, but he thought Mr. Dewhurst would bear him out if he said that it would require 30,000 ft. or 40,000 ft. of sediment to account for even the Tertiary sequences in Burma alone. While cases existed in which the actual thickness measured was greater on the margin of the geosyncline than in the centre that was not always the case. A thousand feet of coarse arenaceous sediment from the point of view of time of deposition was equivalent to, say, 100 ft. only of sediment of another character such as a fine shale, and for the purpose of constructing a purely diagrammatic section one was justified in making allowances for the difference. Mr. Lees in the next place had taken exception to the imaginary mountain chain depicted in diagram 3, Fig. 4, the main reason being that he (the author) had ignored the various phases of mountain building. It was true that one does not get a geosyncline gradually filled up with a nice lot of sediment and then the whole buckled at once. In all the larger cases it took place in stages. There was erosion, and consequently unconformities developed, then further sedimentation and a repetition of the process. He ventured to say that a diagram constructed from such an actual case would merely vary from diagram 3 of Fig. 4 in showing those unconformities. The other point to be embodied would be merely the question of the thickness of the sediments. Mr. Lees had also taken exception to diagram 3 of Fig. 5, and, rather to his astonishment, had quoted a lecture which was given to the Geological Society regarding the structure of the Pamir knot. Personally, he would take that as being a very special case, where two great folded arcs approached one another, rather than a typical case, so that he considered the example given by Mr. Lees in support of his argument was a very unfortunate one. He definitely held to the truth of the diagram in Fig. 5 as being characteristic of a number of mountain chains. He was glad Mr. Lees was satisfied that they knew all there was to be known about the structure of the Alps. In view of the diverse opinions that were held by those who wrote on the subject, personally he would hesitate to say that such was the case. Whilst it might be true that in certain cases there had been overthrusting from two directions, north and south, he differed from Mr. Lees in considering that it was not necessary, and he would go so far as to say that he did not think it was typical. The great work of Staub on the Alps stressed thrusting from the south, and Staub's sections agreed closely with the diagrammatic ones given in the paper as typical. Then Mr. Lees had taken exception to some of his generalisations and had

himself plunged boldly into a realm of speculation which was still more dangerous—namely, the question of Palæozoic and Mesozoic climates. Mr. Lees had stated that the climate was the same all over the world in Palæozoic and Mesozoic times. He thought the point was not proven, and he would like to have some evidence for it. There was a possibility that the more primitive organisms which existed in Palæozoic times might not have been so particular with regard to their habitat, and that the differences of temperature might have been just as great from north to south as they were at the present time. He felt that the question of climate in the past was entirely outside the scope of his paper. With regard to the remarks of Mr. Romanes, he expressed his agreement with the writer that the distribution of oil-shale was quite a different matter from the distribution of oil as such, and that a distinction should be drawn between oil-shale and oil-soaked shale, or shales soaked with free oil from other sources. It was undoubtedly a fact that oil-shales occurred amongst continental deposits which were laid down for the most part in small basins on continental masses. With regard to the points raised by Mr. Cunningham Craig, it was as he, the Chairman, had said, the geosyncline was an area of active movement, the movement being downwards until a point was reached when the instability of the structure resulted in the formation of anticlines. He was sorry the Chairman had tackled him on the question of continental drift, because he (the author) thought he had most carefully avoided that dangerous point. It was mentioned in the paper, but he mentioned it in such a way that two other alternatives were allowed to the reader, who could accept any one of the three. He refused to be drawn into any discussion of Wegener's theory or any movements connected therewith. The Chairman was, of course, perfectly correct in the remarks he had made with regard to the existence of the three great movements. There were examples in North America of oilfields associated with them; but, as had been pointed out, 70 per cent. of the oil of the world came from the flanks of Alpine folded ranges, and therefore he directed his attention to a consideration of the Alpine folds. The question of the eastern part of the Burmese basin had cropped up again. As he had spent the greater part of a rather weary nine months in mapping that eastern part in the hope of finding oil and finding none, he agreed with the speaker that it was almost certain that that area had never been petroliferous.

The meeting then terminated.

Liquid Fuels other than Petroleum.*

By A. E. Dunstan D.Sc., F.I.C., F.C.S., and H. G. Shatwell, Ph.D., M.Sc., A.I.C.

Introduction.

This is a singularly inopportune time to discuss the replacement of petroleum by substitute fluids. Whilst the possibility of an early failure of petroleum supplies has been discussed ever since the beginning of the industry, there has been during the past twelve months definite over production in the American fields. The question of continuity of supply is not characteristic of oil alone, but is common to every industry which depends upon a natural raw material. It must be borne in mind, however, that with the progress of technique in winning crude oil, a far higher percentage of production is being obtained to-day than has been possible heretofore, and that means may eventually be found to recover the large ratio of oil remaining in sands which, by present methods, must be considered to be exhausted. Again, it must be remembered that practically every existing oil field has been discovered as the result of surface indications, and some of the seepages have been exploited for many generations. It is clear that a purely accidental phenomenon of this sort cannot characterise every latent source of oil, and now that geological exploration has been supplemented by geophysical science, one must expect a very considerable increase in the location of productive territory.

The position has been admirably summarised by Mr. G. H. Jones in an address to the Chamber of Commerce, Oil City, Pa. (*Oil and Gas Journal*, February 3rd, 1926). Mr. Jones stated that " the U.S.A. has actually in sight 4,500 million barrels of crude—say, six or seven years' consumption. New reserves are being discovered very rapidly, and such discoveries hitherto have not failed. . . . Ten years or even a generation ahead I believe there will be adequate supplies of motor fuel. Our well oil will continue to meet essential requirements, such as motor fuel and lubricants. I believe also it will remain the cheapest source of all these products. If in all this we are mistaken, if well oil fails or becomes an expensive supply . . . we are already in a position to make

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motor fuel from oil-shale and coal. . . . There is little possibility of our supply failing. . . . Well oil supplies cannot fail suddenly. . . . When the supply does begin to dwindle, as eventually it must, the decrease will be extremely gradual, and other motor fuels will come into the market to supplement the gasoline supply."

The phenomenal growth of the oil industry has recently been described by Sir Frederick Black ("Economics of the Petroleum Situation in British Dominions"). "In the year 1880 the total (production of crude oil) did not much exceed four million tons. Ten years later it was ten millions, and by 1900, the practical beginning of the 'oil age' (*i.e.*, when internal combustion engines were influencing oil consumption), the figures exceeded 20 millions. At the beginning of the war the total was well over 50 millions, and in the first year after the war had reached 76 millions of tons. The next year, 1920, sent the total phenomenally to about 95 million tons. The 1926 figure was probably somewhere near 154 million tons, and 1927, according to a recent estimate by the editor of *Oil News* (London), may show only a moderate increase over 1926."

These facts have served not only to confound the pessimists, but to show that there is not the slightest indication that the civilised world need anticipate a shortage of petroleum in our time.

Nevertheless, despite the continual discoveries of new deposits of crude oil, the rapid increase in production, and especially the recent heavy falls in prices, an intensive search for indigenous liquid fuels in coal-bearing countries has been in progress for some years. Further, important investigations in which the conversion of solid carbonaceous materials into liquid fuels is aimed at have progressed extraordinarily well, and widespread interest is being taken in the commercial possibilities of the discoveries which have already been made. These activities are influenced only to a small degree by economic considerations, because of the abundance and cheapness of petroleum, but have their source almost entirely in political and strategic movements. One of the results of the Great War, as Sir J. Cadman points out, is the realisation of the extraordinary dependence of modern civilisation, more particularly during hostilities, on adequate supplies of fuel for the internal combustion engine, and it is not going too far to state that in current methods of warfare, entire armies, terrestrial and aerial, will be entirely immobilised unless the necessary supplies of liquid fuels are available. These are the factors which explain the feverish attempts of countries lacking in deposits of crude oil to utilise in new ways their reserves of coal, lignite and oil-shale.

At the same time there is an economic factor brought about by the very fact that solid fuels are solid, introducing for this reason the factor of handling costs of the first importance.

The most important mineral asset we possess in this country is coal, the mining and distribution of which find employment for well over a million men. It is a source of energy upon which practically the whole of the country depends, and yet the bulk of its cost is absorbed in medieval methods of handling and transport. If one considers briefly the conditions under which coal is mined, man-handled in the pit, man-handled at the bank, man-handled into trucks and man-handled at the journey's end, it is indeed remarkable that this fuel can be brought to the furnace as cheaply as it is, and it is from this point of view that political considerations should be given due weight.

It may be justifiable at this stage to indulge in, perhaps, vain prophecy concerned with contemplation of the possibilities of rendering solid coal a fluid substance, or energy that can be transmitted through pipes or wires. In brief, one must foreshadow the very definite ideal of the conversion of coal at its source into fluid and energy. Many years ago Sir William Ramsay, in a somewhat Utopian mood, discussed the apparently wild possibility of gasifying coal subterraneously, and, in effect, converting the coal mine into a complete gasification gas works. The idea is not so preposterous as it seems. What one has in mind is gasification of the coal at the earliest moment after its extraction from the bowels of the earth.

If one could imagine a carbonisation process capable of handling the bulk of the output of a given mine, converting the raw coal, after washing, into some form of coke, gas and tar, utilising a portion of the coke in so far as is necessary at the moment as solid fuel and converting the increasingly larger quantity of it into water-gas, there would remain, therefore, for the production of energy, gas and tar. The question of tar disposal will be discussed later in the paper, but a word may now be said relative to the distribution of the gas, which, under this scheme, would be made on a gigantic scale. It is an item of common knowledge that the distribution of any product is a far more important matter finally than its manufacture. This consideration applies with some force to current methods of conveying gas through low pressure mains to our great centres of population. In America, the distribution of natural gas from the wells has assumed immense importance, chiefly because the gas is handled at high pressure, and way-leaves for the lines are easier to obtain. It is in this direction that the railways might obtain a quid pro quo for the loss on the vast amount of mineral transport they cater for to-day.

The existing rail roads offer means of laying pipelines from the colliery districts to the great towns, and the supply of high-pressure gas from the pit to the existing holders offers no very serious difficulties.

Coming now to detail, the major developments in methods aimed at the production of liquid fuels which more or less resemble petroleum, or which may serve as substitutes for certain fractions of crude oil, will be considered. Amongst the substances available for application in this direction are carbonaceous bodies like peat, lignite and bituminous coals, oil-shale, torbanites, and certain types of tropical and sub-tropical vegetation. Of these various raw materials, bituminous and brown coals are of most importance in Europe, and they have provided the basis for notable advances in the technique of synthesis of useful liquid products.

Three distinct lines of attack upon the problem of partially or completely liquefying coal may be discerned. In England, researches have been confined almost entirely to methods of distillation or carbonisation at temperatures substantially lower than those employed in gasworks and coke-oven practice, and the results, both in yields of liquid products and extent of commercial application, have been disappointing. On the Continent, however, and especially in Germany, investigations into the possibilities of utilising coal more rationally have proceeded on a variety of lines, the main object being the complete liquefaction of coal, either by direct hydrogenation or by low-temperature carbonisation, followed by gasification of the coke and subsequent catalytic transformation of the gases into liquid fuels.

Low-Temperature Carbonisation.

The main object of all systems of low-temperature carbonisation of bituminous coal is the production of a solid, smokeless fuel, which may be employed either in domestic fireplaces or in the pulverised state for burning under boilers. The low-temperature or primary tar which is simultaneously obtained can only be regarded as a by-product, and the valuable researches which have been conducted at H.M. Fuel Research Station show definitely that the average yield of such liquid product from suitable British coals is of the order of 16 gallons per ton. It is clear, therefore, that the success of low-temperature carbonisation will depend almost entirely upon the demand for the main product, viz., semi-coke, and that the liquid by-product can supply but a fraction of the fuel oil requirements of this country.

The investigations which have recently been carried out on the chemical nature of primary tar show, further, that this material

cannot be regarded as a complete substitute for petroleum (*cf.* Edwards, *J.S.C.I.*, 1924, 143–156 T: Brittain, Rowe and Sinnatt *Fuel*, 1925, 4, 263-9, 299-307, 337-340: Parrish and Rowe, *J.S.C.I.*, 1926, 100-106 T: Fisher and collaborators, "Conversion of Coal into Oil," 1925).

Naturally, the composition of low-temperature tar varies with the coal from which it is derived and the method of distillation adopted. If external heating of the retorts is practised, the tar may be expected to contain a higher proportion of low-boiling constituents than when the coal is distilled in a current of heated gases. On the other hand, the tar prepared by the latter method, other conditions being the same, will be less "cracked," and may possibly contain fractions from which lubricating oils may be extracted.

However prepared, primary tars consist chiefly of hydrocarbons and phenols or tar acids; sulphur, nitrogen and other oxygenated compounds are present in variable but small proportions. From the liquid fuel point of view the hydrocarbons are the most valuable constituents, since the other components of the tar possess low calorific values or undesirable properties. Further, only the hydrocarbons can possess properties which will render them suitable for employment in petrol engines or as illuminants and lubricants. Consequently, unless the tar is to be used as boiler or diesel engine fuel (*cf.* O. Schertel, J. Gusbeleucht, 1918, **61**, 493–5), the tar acids and bases must first be removed.

The extent to which phenols occur in primary tars differs considerably. Thus, tars examined by Brittain, Rowe and Sinnatt, Parrish and Rowe, and Fisher have been found to contain 30·6 per cent., 33·84 per cent. and 50 per cent. respectively of constituents soluble in caustic soda, whilst the authors have dealt with tars containing 21 per cent. and 26 per cent. respectively of tar acids. Consequently, it would appear that approximately a third of the low-temperature tar which may be thrown on the market will be composed of substances applicable only for use at the moment as boiler fuel, or possibly for combustion in diesel engines, though the insolubility of tar acids in petroleum will not commend their utilisation for the latter purpose. The only outlet for phenols at the present time is in the manufacture of synthetic resins and disinfectants, and until some alternative use for them is found the commercial value of low-temperature tar will be of a low order. Their chief potential value lies in the fact that they differ from aromatic hydrocarbons only in containing oxygen, and that if this element can be eliminated by some method of reduction, substances of technical value will be obtained. Hitherto, the conversion of phenols into aromatic hydrocarbons has not proved practicable

on the industrial scale, but the increasing knowledge of catalysts may shortly lead to a solution of the problem.

The material remaining after elimination of phenolic and basic substances, and constituting, on the average, two-thirds by volume of the crude tar, is characterised by its high content of unsaturated hydrocarbons and of pitch. It is more closely related to the product derived by cracking a heavy petroleum fraction than to a natural crude oil, and its treatment must be modified accordingly. The pitch which remains as a residue of distillation either of the crude tar or its hydrocarbon components is of little use for employment for road-surfacing or briquetting purposes, but its low ash content may render it of value in certain specialised industries. The remaining hydrocarbons are largely naphthenic in type, though paraffins and wax are usually present, with occasional traces of benzol and other aromatic hydrocarbons.

Examination of tars by methods of distillation are frequently based on coal-tar practice, and the results so obtained are difficult to collate. Parrish and Rowe (*loc. cit.*) give the following results of distilling a primary tar produced by the carbonisation of coal from the Pooley Hall seam in vertical retorts, the yield of tar being about 14 gallons per ton of coal.

Fraction.	B.P.	% of tar by weight	d_{15}^{15}	Tar acids % vol.	Bases % vol.	Neutral Oil % vol.
1 ..	up to 170° C. ..	2·79	0·850	10·2	1·7	88·1
2 ..	170° C–230° C. ..	20·45	0·949	45·9	1·0	53·1
3 ..	230°–270° ..	14·35	0·981	49·5	3·5	47·0
4 ..	270°–310° ..	12·69	1·003	37·6	2·3	60·1
5a ..	310°–335° ..	10·07	1·132	33·2	2·4	64·4
5b ..	Residue ..	36·06	—	—	—	—
	Loss	1·30	—	—	—	—

Sinnatt and King (*J.S.C.I.*, 1925, 413–424 T) have made an attempt to separate a low-temperature tar arbitrarily into marketable products, as follows :—

Refined Products per Ton of Coal.

		% on Crude Tar
Refined spirit from Gas	= 1·40 galls.	8·6
Light naphtha or spirit to 160° C. ..	= 1·2 „	7·3
Heavy naphtha to 200°	= 0·7 „	4·3
Burning oil to 270° C.	= 1·8 „	11·0
Gas oil to 300° C.	= 0·8 „	4·9
Light lubricating oils to 360°	= 0·7 „	4·3
Tar Acids, 190° to 310°	= 2·7 „	26·6
Pitch	= 50 lbs.	37·0 (approx.)

The above figures, which are typical of most British primary tars produced in externally heated retorts, indicate high contents

(exceeding 50 per cent.) of tar acids and pitch, and low contents of intermediate oils. The spirit fraction, though highly unsaturated in type, has been found by the authors to be amenable to refining by the conventional sulphuric acid and sodium hypochlorite methods, giving a refined spirit which is water-white, stable to light, and of sweet odour. The most valuable property of this product, however, is its ability to resist detonation in internal combustion engines, making it a useful agent for blending with those spirits which are prone to knock in the engine.

Very little is known about the kerosine fraction of primary tars except that its refining will necessarily be of a drastic nature, and will be accompanied by heavy losses of material.

Finally, the possibilities of obtaining lubricating oils from low-temperature tars are very much in dispute. Good lubricants are claimed to have been produced from coal tar in Germany during the war (*J.S.C.I.*, 1919, 331 R), and Fischer and Gluud (*Ges. Abhandl Kennt. Kohle*, 1917, 114–142; *J.S.C.I.*, 1919, 492 A) obtained viscous oils having a deep orange colour and a green fluorescence from primary tars prepared by distilling coal in a revolving retort. They state that it is essential to avoid excessive heating of the coal and to remove the volatile products rapidly from the distillation chamber. The viscous oils, which were generally heavier than water, were obtained in yields of 15·2 per cent. of the tar from a bituminous coal, 10·0 per cent. from a gas-coal tar, and 11·3 per cent. from a producer tar. Recently, primary tar prepared from a Shirebrook bastard cannel in the Laing and Nielsen rotating, internally heated retort was found to contain 15 per cent. of a lubricating oil base, not more than 4 per cent. of which was spindle oil (*Engineering*, 1927, **123**, 665–6). The L. and N. plant, in which the coal is heated by a current of hot producer gas, satisfies the conditions of coal treatment previously elaborated by Fischer and Gluud. In view of the importance attached to the necessity of manufacturing lubricating oils from indigenous materials during times of national emergency, it would be of great interest to learn the effects of distilling bituminous coals, as opposed to cannel, in the L. and N. or other process.

An alternative method of dealing with low-temperature tars has been investigated by the authors. Two tars of different origin, but both derived from British bituminous coals, were subjected to thermal decomposition or "cracking," after elimination of low-boiling constituents. It was found that higher temperatures than is customary with mineral oils were required to effect any extensive degree of cracking, that the tar acids were decomposed into gas, pitch and simpler phenols, but that their decomposition did not give rise to measurable yields of hydro-

carbons, and that the production of pitch or coke was of a high order. The crude cracked spirit proved to be readily refined, and, examined in an experimental engine, was found to possess an anti-knock "benzol" equivalent of about 50 per cent.

Hydrogenation of Coal.

The liquefaction of coal by reduction was first effected by Berthelot in 1869, but the first attempts to transfer the process to the industrial scale were carried out by Bergius about 1913–1914. Berthelot employed hydriodic acid as his reducing agent, whereas, of course, Bergius uses hydrogen itself under extremely high pressures. Comparative secrecy has been maintained relative to the exact types of coal experimented with at Mannheim, and the most important conditions of the operation, viz., temperature and time of reaction, can only be inferred by outside observers. It is known, however, that hydrogen pressures of the order of 150 to 200 atm. are necessary, and that the optimum temperature conditions vary for each coal, but are almost invariably within a range of a few degrees. The yields of liquid materials depend partly upon the coal and partly upon the conditions of experiment. Bergius himself stated at the recent International Conference on Bituminous Coal at Pittsburg that from 40 per cent. to 70 per cent. of oil may be produced by treating suitable coals under 200 atm. hydrogen pressure and at 420° C. Also, from an average bituminous coal, 15 per cent. of motor spirit, 20 per cent. of middle oil, 6 per cent. of lubricating oil, and 8 per cent. of fuel oil can be obtained. Sinnatt and King (*J.S.C.I.*, 1925, 423 T), after a description of the intermediate scale plant at Mannheim, state that "certain British coals have been transformed" in this apparatus "into liquid and gaseous products to the extent of 85 per cent. by weight at the expense of hydrogen equal to 10 per cent. by weight of the coal." The liquid product, on distillation, gave the following data :—

Initial boiling point	60° C.
60–150°	15 per cent.
150–200°	25 „
210–230°	30 „
Residue	30 „

"The residue is composed of heavy hydrocarbons resembling pitch. The fraction 210°–230° obtained in one experiment contained approximately 20 per cent. of phenols."

There is no doubt, therefore, that the technical validity of the Bergius process is established, and that from one ton of raw coal an average of 140 gallons of liquid product may be anticipated.

Very little, again, is known about the nature of this new chemical raw material. That it contains substantial proportions of light

hydrocarbons is shown by the above quotations and by the work of Ormandy and Craven (*J.I.P.T.*, 1926, 77–78). The latter found that the spirit boiling below 160° C., equal to 10 per cent. by weight of the coal (30 gallons to the ton), contained 3·1 per cent. of unsaturated hydrocarbons, 7·5 per cent. aromatic hydrocarbons, 47·0 per cent. paraffins, and 42·4 per cent. naphthenes.

At the same time, however, the crude product is rich in tar acids and pitch. It appears probable, therefore, that Berginised coal is closely related to low-temperature primary coal-tars, and that its treatment will warrant special investigation. The presence of lubricating oils (quoted by Bergius) is of some interest. It is extremely unlikely that coal-oils obtained at temperatures exceeding about 430° to 450° C. will contain bodies comparable with petroleum lubricating oils, because of their tendency to undergo thermal decomposition. It appears probable that two main reactions occur when coal is hydrogenated: (*a*) absorption of hydrogen and the formation of a viscous mass which may be termed the primary product. (These products have been obtained by Graham, Shatwell and Bowen in hydrogenating coal at temperatures of 400°–425° C.) (*b*) Decomposition and hydrogenation of the primary product at temperatures exceeding 430°–450° C., with the formation of pitch and light products at the expense of the intermediate fractions.

Whatever be the nature of the chemical reactions involved in the "berginisation" of coal, the discovery of Bergius is of profound scientific interest, because it provides a new method of investigating the composition of coal. From the industrial point of view, its future depends upon a large number of economic factors, not least amongst which is the cost of hydrogen. When it is realised that as much as 30,000 cub. ft. of hydrogen may be required to liquefy one ton of coal, and that hydrogen of the required degree of purity cannot be manufactured by present methods for less than 2s. per thousand cub. ft., the importance of this factor becomes obvious. For this reason, Bergius has concentrated on methods aimed at recovering the hydrogen, which is contained in the form of methane, ethane, etc., in the effluent gases of his process. The first attempts consisted in scrubbing the gases at low temperatures and high pressures with liquids, such as ether, tar or petroleum oils, in which hydrogen is only slightly soluble, whereas the impurities dissolve with ease (G.P., 394,497, 1921). More recently, however, a process has been evolved in which the hydrocarbons, which may be present to the extent of 30 per cent. in the waste gases, may be decomposed and the hydrogen recovered. By this means it is possible to obtain from the coal itself a considerable proportion of the hydrogen required for its reduction. The gas, after removal of light spirit and

sulphuretted hydrogen, is mixed with steam and heated to about 1,100° C. The steam and hydrocarbons interact at this temperature, producing hydrogen and carbon monoxide, as follows:—

$$C.H_4 + H_2O. = CO. + 3H_2.$$

The resulting mixture of carbon monoxide and hydrogen is then cooled somewhat, mixed with a further quantity of steam, and passed through a catalyst, such as iron oxide, heated to 300° to 500° C. This treatment converts the carbon monoxide into the dioxide, which may be eliminated in one of many simple ways.

$$CO.H_2O = CO_2H_2.$$

The final result is that from one molecule of methane and two molecules of steam four molecules of hydrogen are obtained.

Other recent developments of an important nature are concerned with the use of catalysts to accelerate the reactions between coal and hydrogen (I.G. Farbenindustrie A.G., English Patents 247,582–4, 251,264, 272,829, 1926). The catalysts disclosed are nitrogen in the combined state, molybdenum, chromium, tungsten, and their compounds, or combined sulphur.

Gas Reactions.

The third method of producing synthetic fluid fuels, and one which aims at the complete liquefaction of the coal, has as its initial stage the production of water-gas by the interaction of steam and coke. Water-gas, which has the approximate composition hydrogen 50 per cent., carbon monoxide 40 per cent., carbon dioxide 5 per cent., nitrogen 5 per cent., is used extensively as a fuel, but the reactivity of its main components has been the subject of much investigation. Thus Sabatier in 1902 found that carbon monoxide could be reduced to methane by the action of excess of hydrogen in presence of nickel at 230° to 250° C., and much subsequent work was undertaken on these lines with the object of eliminating carbon monoxide from, and increasing the illuminating power of, coal gas.

Methyl Alcohol.

The investigations, however, took a new turn when the Badische Anilin und Soda Fabrik (B.A.S.F.) discovered that liquid products resulted by contacting a mixture of carbon monoxide and hydrogen with a variety of catalysts at temperatures of the order of 400° C. and pressures of 120 atmospheres (G.P. 293,787, 1913). This discovery was the prelude to a series of researches which have shown that a large variety of products can be obtained when carbon monoxide is caused to react with hydrogen under varying

temperatures and pressures and in presence of different catalysts. Work by the Badische Company was interrupted, partly by the war, and partly by the fact that the heterogeneous nature of the products was thought to militate against the commercial exploitation of the new process. The results, however, laid down the lines on which later researches should be carried on, the objective being the production of methyl alcohol. Simultaneously, Patart, in France, was investigating the same series of reactions, and caused to be erected in 1922 a small-scale plant in which the required material was successfully prepared, while the B.A.S.F. a little later began the actual manufacture of methanol on a commercial scale. On the other hand, Fischer and Tropsch, at the Institute of Coal Research, Mulheim-Ruhr, returned to the reaction originally investigated by the B.A.S.F., their object being the preparation, not of methyl alcohol, but of substances which would serve as substitutes for motor spirit. As a result of their experiences in this work, the latter investigators proceeded a step further, and actually synthesised a product which consisted entirely of hydrocarbons—gaseous, liquid and solid—and virtually established the possibility of synthesising petroleum, starting with coal and water.

The reactions which water-gas can be made to undergo are, therefore, many and varied, and since the work is only yet in an early stage, the ultimate possibilities are of extreme interest.

The processes of the Badische Company (E.P.s 227,147, 228,959, 229,714, 229,715, 1923; 231,285, 237,030, 240,955, 1924), and of Patart (*Compt. Rend.*, 1924, **179**, 1330–2; *Chem. et Ind.*, 1925, **13**, 179–185: see also Lormand, *J. Ind. Eng. Chem.*, 1925, 430–2) are not radically dissimilar in principle. Whilst the former prefers as catalysts metallic oxides or their mixtures, especially the oxides of copper, silver, zinc, lead and cadmium, the latter specifies all the metals, oxides or salts which are known to favour oxidation or hydrogenation, such as the oxides of vanadium, chromium and zinc, preferably mixed with copper, and such salts as chromates, manganates, vanadates, molybdates and tungstates. In both cases metals of the iron group—iron, nickel and cobalt—must be rigorously excluded, the B.A.S.F. even carrying this restriction so far as to preclude such metals as components of any hot portion of the plant, whilst, also, the reacting gases contain hydrogen in excess (1·5 to 2 volumes of hydrogen to one volume of carbon monoxide). With regard to other experimental conditions, the B.A.S.F. carefully purifies the gases before interaction, whereas Patart effects only partial purification in a single scrubber, whilst the latter works at pressures of 500 atmospheres and the Badische process at about 200 atm. Temperatures in each case approximate to 400° or 420° C.

Patart's experimental plant gave a conversion of gas to liquid of from 8 per cent. to 10 per cent. with one passage of the gas, and he further estimated that 100 litres of methyl alcohol can be produced from 300 cu. metres of water-gas at a cost of 25 to 30 gold francs.

Fischer, who has carried out experiments on the same lines, found that equal volumes of carbon monoxide constituted the most suitable mixture to be operated on, that zinc oxide was the best catalyst, and that iron, nickel and cobalt were detrimental to the course of the reactions. At temperatures and pressures of 400° C. and 150 atm. respectively the yield of methanol was almost theoretical, the crude product containing only traces of acids, and containing 93 per cent. of constituents boiling below 80° C. The zinc oxide was still pure white, after use extending over some weeks.

Synthetic methanol is now a recognised article of commerce, and finds extensive use in many directions; as a motor fuel, however, it is not a success. Its calorific value is low, and its incapability of resisting pinking in the engine will prevent its adoption, for this purpose, on any large scale.

Synthol.

It was probably for these reasons that Fischer and Tropsch began to investigate the original Badische process by which liquid hydrocarbons were stated to have been obtained. In their first experiments, they repeated exactly the conditions specified in German Patent 293,797, 1913, Example 2, *i.e.*, purified water-gas containing an excess of hydrogen was passed at 400° to 420° C. and 90 atm. pressure over a catalyst consisting of alkali and zinc carbonate on pumice, but the original gas emerged practically unchanged, and not a trace of oil was obtained (*Conversion of Coal to Oil*, p. 221). Subsequently it was found that, whereas, in the preparation of methanol, iron group metals are definitely inhibitory, their presence if other products are required is essential, and at the same time the simultaneous use of strong alkalis is necessary. In short, if water-gas is passed over a catalyst composed of iron borings impregnated with a strong base in the form of a hydroxide or carbonate, the temperature being 410° C. and the pressure 150 atm., the reaction product consists of watery and oily layers. The oily components, known as synthol, contain no hydrocarbons, but are composed of a complicated mixture of higher alcohols up to C_{18}, of fatty acids to C_8, and of aldehydes, ketones and esters. The proportion of this product can be increased by using strong bases, *e.g.*, rubidium carbonate, by having a greater thickness of catalyst, and by operating upon a gas mixture containing a less proportion of hydrogen.

The pressure should be as high as is conveniently possible, and not less than 70 atm. Although finely divided iron, in conjunction with alkali, is an effective catalyst, iron borings or turnings are preferable in that their activity is more prolonged and carbon deposition is reduced to a minimum.

A single passage of the gas is not sufficient, so that a continuous plant has been constructed in which the water-gas can be continually re-circulated under pressure, the condensable products being removed at each successive stage. In the earlier experiments, Fischer and Tropsch converted 27 per cent. of the heat value of the gas into synthol, but it is believed that conversion has now attained to 46 per cent.

Synthol is described as a light yellow liquid having a calorific value of 7,540 to 8,200 calories per k. gram. Its specific gravity is 0·8289. When chilled it begins to thicken at $-30°$ C., and is completely solid at $-90°$ C. Since, further, 97 per cent. is volatile below 200° C., Fischer and Tropsch have proposed its utilisation as motor spirit, and have described its performance in a road test using a motor cycle carrying two persons. The mileage obtained showed that synthol was equal, if not superior, to benzol (*loc. cit.*, p. 247).

The manufacture of synthol on a large scale was undertaken some time ago in Germany, but its tendency to knock and other properties have proved it to be unequal in value to petroleum spirits. Nevertheless, the results may be of great value in subsequent years.

Synthetic Petroleum.

The work briefly described in the preceding sections indicates that catalytic reduction of carbon monoxide at high pressures leads to the formation of oxygenated compounds, whereas at atmospheric pressure and a high temperature methane is the sole product. Investigating the latter reaction more thoroughly, Fischer and Tropsch elicited the fact that it is possible to obtain products fundamentally different from those previously obtained, and that the reduction of carbon monoxide can be effected at quite low temperatures. For example (*Brenn. Chem.*, 1926, **7**, 97–104), they describe the formation, from water-gas, of homologues of methane, consisting of a series of gaseous, liquid and solid hydrocarbons, the heaviest of which have 70 carbon atoms in the molecule. The most effective catalysts are iron, cobalt and nickel, mixed with a small proportion of the oxide of the same metal, though many other metals or oxides may be employed. The optimum temperatures are 300° C. with iron, 270° C. with

cobalt, and 160° C. with nickel. Although it is usual to operate with carefully purified water-gas, successful results have been obtained with mixtures corresponding to generator gas and furnace gases.

The product, as stated above, consists of water, carbon dioxide and hydrocarbons, the lighter portions of the latter passing away from the catalyst tube to condensers and scrubbers, whereas the heavier fractions, including solid hydrocarbons, collect in and around the catalyst and eventually prevent it from functioning. Consequently the contact material has to be steamed periodically. It has also been found that the addition of a little alkali to the catalyst increases the molecular weight of the resulting hydrocarbons. Thus, already, it is known that the boiling range of the product can be varied.

The researches on the synthesis of hydrocarbons have not yet progressed to the extent that an opinion can be expressed as to the probable commercial value of the chemical reactions involved. Their scientific importance, however, is enormous, and the results which have accrued in a relatively short space of time in working with such an apparently unpromising material as water-gas are encouraging, and must stimulate further researches in the realm of fuel technology.

The Conversion of Coal into Oil by the Bergius Method.*

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The enormous increase throughout the world during the last few years in the use of motor vehicles depending upon the internal combustion engine, has resulted in a demand for motor spirit of varying grades which threatens to shorten considerably the life of known oil resources. Moreover, the demand for fuel oil has very considerably increased, consequent upon its widespread utilisation for steam raising and in Diesel engines. The many advantages possessed by liquid fuels over those in solid form, notably higher calorific values, greatly superior thermal efficiency of internal combustion engines and absence of ash and ease of transport, make worth while the payment of anything from three to five times as much for heat units in the form of hydrocarbon oils as for the same amount of latent energy in the form of raw coal. How long such a state of affairs will continue will, no doubt, depend upon the application of other forms of fuel to the needs of motor transport. At the present time liquid fuel is certainly the most convenient source of energy, and for countries possessing no natural oil supplies its production from coal at a reasonable cost is, of course, a matter of great national importance. All but a small proportion of the total quantity of liquid fuel used in the British Isles is imported. This relatively small amount is produced either by low temperature or high temperature carbonisation—both being processes in which the oil is simply a by-product—whether it be from coke ovens or gas works. In the Bergius process for the conversion of coal into oil the position is reversed—the liquid fuel is the chief product whilst the gas and coke are by-products.

It is well known that the estimated coal resources of the world far exceed those of oil. In this country, in particular, the natural oil resources are, comparatively speaking, negligible. The probable coal resources of the United Kingdom were estimated by Prof. H. S. Jevons in 1915 at 197,000 million tons—the estimate being

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based on a limiting depth of 4,000 feet, and all seams one foot and upwards in thickness. Taking 250 millions as an annual output, exhaustion of coal supplies might be expected to take place in 800 years, making allowance on one hand for losses in mining, and on the other for coal worked below the 4,000 feet line—*not* an impossibility. The coal resources of this country, however, are said to form only 2·6 per cent. of the estimated world resources, and it behoves us as a nation, therefore, to utilise to the full the latest possibilities of our coal seams both in mining and by the subsequent treatment and utilisation.

According to the published results of the Bergius process, about 50 per cent. of the coal may be converted into oil. Reckoning the annual consumption of motor spirit to be of the order of 2½ to 3 million tons, and that of fuel oil an equal amount, the present liquid fuel needs of the country would be met by treatment of approximately 15 million tons of raw coal. Obviously, therefore, hydrogenation cannot be a panacea for the ills of the mining industry, since only about 6 per cent. of the total amount of coal raised would be required in the process. Nevertheless, as we hope to point out later, the process, if economically feasible, should bring greater prosperity to those coal fields possessing seams of coal readily amenable to hydrogenation.

Without disparaging in the slightest the plant evolved through the hard work of Dr. Bergius and his colleagues, it is only natural that improvement may be expected and simplification of design result from further research. That hydrogenation of coal will have a future, we believe there is no doubt—though its scope will be restricted within certain limits.

Our object in this paper is, however, to put before this Institute the present position so far as we see it, and to lay before members the results of laboratory experiments. These, we trust, will be of service in showing which types of coals of Great Britain are most readily amenable to high pressure hydrogenation according to the method of Bergius.

The Bergius method appears to have been evolved as a result of experiments made by the inventor, on the formation of artificial coal from cellulose, peat, lignite, etc. This material was prepared by heating in an autoclave the cellulose, in presence of water, at temperatures up to 340° C., and was found by Bergius to reach the same ultimate composition irrespective of temperature, the time taken to produce this material of constant composition, "endkohle," being, however, immensely greater the lower the temperature of the reaction. For example, the product obtained by treatment of cellulose under water for 21 hours at 340° C. apparently possessed the same composition as the product of 22½

hours at 310° C.—the "endkohle" in each case possessing approximately 84 per cent. carbon. Bergius discovered that by treatment of this "endkohle" with hydrogen under high pressure at 400° C., about 70 per cent. could be converted into an oil soluble in benzene and very similar to petroleum. His theory (though perhaps not correct) that natural coals are similar to this "endkohle" in chemical constitution prompted him to carry out similar experiments with natural coals. These investigations were followed by a series of patents taken out in different countries in the years 1913 and 1914, the first English one being granted on August 1st of the latter year. This stated that the invention "relates to the treatment of coal and other solid carbonaceous material of vegetable origin formed by natural or artificial carbonisation to produce directly by chemical conversion of the carbonaceous material liquid hydrocarbons or hydrocarbons of a low melting point, and consists in subjecting carbon or the like to the action of hydrogen at pressures above 10 atmospheres and preferably exceeding 100 atmospheres, and at an elevated temperature less than 600° C. In these conditions the hydrogen reacts directly with coal, which is transformed *without destructive distillation* and substantially without formation of uncondensable gases into valuable hydrocarbons which are either liquid at ordinary temperatures and pressures or which have a low melting point. The working temperature is preferably between 300° and 500° Cent. In place of hydrogen, gases containing free hydrogen, such as water gas, could, of course, also be used. . . ." The presence of a solvent is considered (according to the specification) to be advantageous, and hydrogenation may also be combined with carbonisation by subjecting wood, peat or other vegetable matter to heat treatment in a hydrogen containing atmosphere at high pressure.

The oil most desirable from the point of view of internal combustion engines should contain carbon and hydrogen only, in the ratio of approximately 8 : 1. Since coal consists in the main of carbon, hydrogen and oxygen, with smaller quantities of nitrogen and sulphur, it is obvious that for conversion to a hydrocarbon oil, the oxygen, nitrogen and sulphur must be removed. At the same time a considerable amount of hydrogen must be added in order to lower the C : H ratio in coal from about 16 to about 8 in oil. Bergius has claimed that this can be done by high pressure hydrogenation—that the oxygen is removed as water, the nitrogen as ammonia, and the sulphur as sulphuretted hydrogen, and that nothing but the ash of the coal will be left. Assuming this possibility, hydrogen will have to be taken up by the coal to the extent of approximately 6 per cent. of its weight. That coal can be made to react with hydrogen and become converted into oil, has

been proved by the classical experiments of Berthelot in 1869. This celebrated scientist treated coal with hydriodic acid in sealed glass tubes in a furnace heated to 270° C., and obtained as a result of the reaction oils in property comparable with a natural petroleum. This method is obviously not a practical proposition, and it is to Dr. Friedrich Bergius and his colleagues that credit is due for the production, 40 years later, of a plant for the conversion of coal into oil on a commercial scale. Nevertheless, the hydrogenation of coal is not an easy matter, and the results of the work of Bergius and his collaborators, first published in 1925 (Zeit. des Vereines deutscher Ingenieure Band 69, Nr. 42/43, 1925), have also indicated that the complete conversion of the coal, leaving nothing but ash, is a more or less practical impossibility.

The molecular complex of which coal is formed would not appear to be readily amenable to direct reduction by hydrogen unless molecules of the latter are present in very great numbers, and the coal is raised to a temperature at which decomposition commences. These conditions are realised by subjecting the coal to treatment at a temperature of about 430° C., and a hydrogen pressure of over 100 atmospheres.

Although carbon and hydrogen do not combine to any appreciable extent at temperatures below 1,000° C., coal and that gas react at temperatures in the neighbourhood of 400° C.—a reaction which, according to Bergius, is uninfluenced by the presence of catalysts.

Bergius's first experiments (in 1913 and 1914) were carried out in a glass-lined metal bomb, the vessel being filled with hydrogen, after introduction of the coal, to a pressure of 100 atmospheres, and then heated to between 350° and 400° C. After several hours' treatment the vessel was allowed to cool, and when opened was found to contain about 15 per cent. of the original weight of coal unchanged, and a considerable percentage of distillable oil.

In subsequent experiments Bergius employed rotating autoclaves in order to obtain more satisfactory mixing of the gaseous and liquid phases during the reaction. The Bergin Actien Gesellschaft, formed to test thoroughly Dr. Bergius's methods for the conversion of coal into oil, is perhaps rather unique as a research organisation. Formed in 1914 with a capital of one million marks and a large staff of scientific and technical workers, this company had for its object the development of the Bergius patents into a sound working proposition. Owing to the War, however, from 1914 until 1921 little progress seems to have been made in the hydrogenation of coal, although considerable technical skill was achieved in the manipulation of large scale apparatus for the hydrogenation of oil.

In 1921 a considerable amount of research was undertaken and new works were equipped and fitted so that an extensive series of experiments with various kinds of coal could be carried out under different conditions. Towards the end of 1922, a plant capable of treating one ton per day was put into working operation, and since that time, treatment of numerous samples of coal from different sources has been carried out.

In 1925 an important paper was read by Dr. Bergius before the Vereines deutscher Ingenieure, giving a detailed survey of the work done by him and his colleagues. Table I gives the results of a number of tests quoted by Bergius—about 25 per cent. of the latter's results being included in our table.

According to Bergius, the amount of insoluble coal substance first diminishes with rising temperature, then reaches a minimum value, and with further rise of temperature increases again. The maximum value of decomposition varies with the nature of the coal, and must be ascertained for each sample. In the case of many of the experiments quoted in the table, hydrogenation was carried out by treatment of the coal in the presence of oil as a distribution medium.

Some of these coals have been tested in the 40 litre bomb as well as the 2 litre apparatus. This table gives the proximate analysis of the coal in addition to the oil yield resulting from hydrogenation. The amount of residue, which Bergius recognises as being different from the original coal, varies with the working conditions as well as with the type of coal used. From a study of Table I there appears to be little connection between the volatile matter and the yield of oil. The latter is shown to vary from 40 per cent. to 70 per cent. of the pure coal substance.

Certain brown coals are stated by Bergius to leave only 1 per cent. of coal residue, whilst for gas flame (long flame, non-caking) coal the residue free from ash amounted to 10 per cent. Flame coals, he considers, give 15 per cent. insoluble. With increasing carbon content the yield of oil decreases. The use of a distribution medium—oil produced by a previous hydrogenation—is advocated by Dr. Bergius. The hydrogenation is followed by means of the pressure-temperature curve, upon the importance of which Bergius lays great emphasis. This curve, he considers, is characteristic for every kind of coal, and important deductions can be made from the curves in regard to the course of reaction of the coal under treatment.

The pressure rises with temperature to a certain point, after which a drop occurs. This is, of course, the result of absorption of hydrogen by the coal. On cooling, the pressure falls to a figure considerably below the original cold charging-pressure, in spite of the fact that

TABLE I.

Results of Hydrogenation by Dr. Bergius of coals from various sources, treated in either 2 litre or 40 litre bombs.

Coal No.	Expt. No.	Proximate analysis of moisture-free coal. Ash.	Pure coke.	Volatile matter.	Volatile matter in the pure coal.	Results. Oil yield as % of Crude coal.	Oil yield as % of Pure coal.	Residual coal as % of Crude coal.	Residual coal as % of Pure coal.
		Rhenish Westphalian Bituminous Coal.							
201	1553	5·3	58·0	36·7	38·7	45·7	50·8	18·8	21·1
205	1509	5·0	65·2	29·8	31·4	39·0	41·0	25·4	27·0
206	1478	31·6	49·5	18·9	29·4	36·4	55·8	10·3	15·7
211	1543	7·7	82·6	9·7	10·5	40·0	44·0	40·7	44·5
215	1586	6·2	70·3	23·5	25·0	50·5	54·2	23·0	25·0
217	1567	7·4	73·1	19·5	21·1	45·0	48·6	32·0	34·6
207	1570	6·3	65·8	27·9	29·4	52·2	56·0	22·2	23·8
209	1534	4·0	70·0	26·0	27·0	42·3	44·0	27·0	28·0
213	1550	9·5	75·5	15·0	16·6	35·7	37·8	26·3	29·5
		Other German Coals.							
220a	1682	9·3	67·7	23·0	25·4	40·0	45·0	27·6	30·0
224	1777	18·3	66·7	15·0	18·4	41·2	51·2	22·0	28·4
221	1789	24·0	54·5	21·5	28·3	43·0	60·0	13·3	18·5
232	389	11·0	50·5	38·5	43·3	56·0	63·0	3·2	3·6
231	1926	34·5	37·0	28·5	43·5	42·8	66·0	3·5	5·1
		Foreign Coals.							
4	1354	28·9	43·3	27·8	39·0	31·5	44·2	27·3	36·4
103b	1466	9·8	54·2	36·0	40·0	56·3	65·4	8·0	9·3
105c	1598	9·5	69·2	21·3	23·5	48·7	55·6	12·9	14·7
104a	1467	5·0	64·0	31·0	32·6	51·0	53·8	17·5	18·4
104c	1471	7·5	73·5	19·0	20·5	43·0	46·8	22·2	24·1
106b	1537	15·7	54·7	29·6	35·1	50·6	57·5	13·9	15·8
108b	1490	4·0	56·0	40·0	41·7	49·8	51·4	18·2	18·8
109a	1518	5·3	59·5	35·2	37·2	51·5	58·0	13·4	15·1
110a	1517	4·8	59·0	36·2	38·0	51·8	55·3	17·5	18·7
111b	1530	8·5	53·6	37·9	41·4	50·8	57·0	13·9	15·6
112a	1549	11·0	49·5	39·5	44·4	57·2	64·8	13·1	14·8
112c	1555	7·1	59·5	33·4	36·0	50·0	54·5	15·7	17·1
116	1674	44·0	37·0	19·0	34·0	29·0	53·0	8·1	14·7
304	1628	5·0	63·5	31·5	33·2	49·0	51·0	17·4	18·3
305	1645	4·5	63·5	32·0	33·5	58·0	61·0	11·3	11·9
308	1633	14·8	43·6	41·6	48·8	56·0	66·0	11·0	11·7
312	1883	7·6	60·7	31·7	34·3	60·0	65·0	13·4	14·5
402	1853	12·2	57·8	30·0	34·2	46·2	53·2	16·2	18·7
407	1920	11·4	70·6	18·0	20·3	28·7	32·4	25·6	28·9
599	522	11·6	40·0	48·4	54·6	54·0	67·7	11·8	14·8
723a	1758	18·3	47·2	34·5	42·2	56·0	71·5	2·8	3·6
801	1369	18·3	43·9	37·8	46·3	47·5	63·0	4·3	5·7
802	1391	2·0	32·0	66·0	67·3	35·0	35·7	18·0	18·4
		Peat.							
303	1655	16·5	32·5	51·0	61·0	37·0	44·0	8·6	10·3

a considerable amount of gas has been formed during the hydrogenation—an amount usually greater than that produced by carbonisation at the same temperature.

For the liquefaction of coal to be a commercial proposition, continuous working is, of course, essential. Dr. Bergius and his

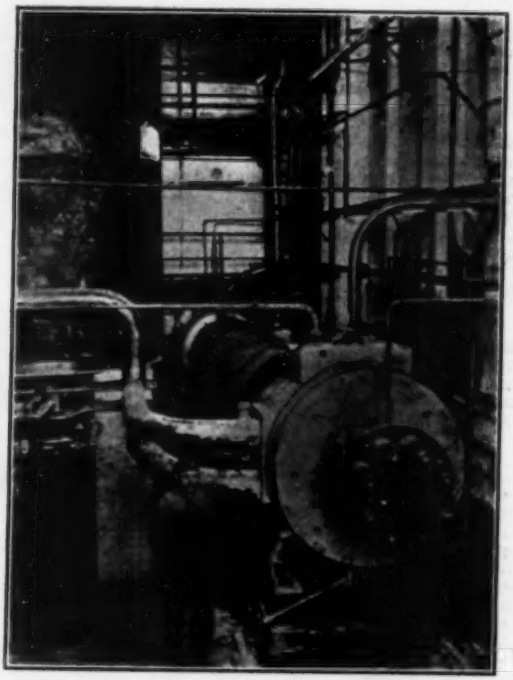

PLATE 1.

Reaction Chamber of a Large-scale Hydrogenation Plant.

collaborators (Dr. Hofsass and others) deserve the highest praise for the way in which they have overcome the many engineering difficulties connected with work on a large scale at pressures in the neighbourhood of 200 atmospheres. It has been pointed out that Bergius considers there is an optimum temperature for each coal, hence a heating arrangement which could be regulated to a fine degree of precision had to be evolved, and this has been made the

subject of a separate patent (E.P. No. 192849). The high pressure receptacle is provided with double walls, and in the annular chamber between these, inert gases (*e.g.*, carbon dioxide and nitrogen) are circulated at a pressure of the same order as that inside the reaction chamber. It is claimed that exact control of the temperature of the reacting materials may be obtained in this way and that recovery of heat from such highly compressed gases can be easily accomplished. The heating medium is raised to a temperature above that of the reaction by passage through a furnace situated in another room.

Plate I illustrates a large hydrogenation plant capable of treating 50 tons of oil per day, which was in working operation in 1922. The autoclave is approximately 30 feet long and 3 feet in diameter. The large pipes shown on the left of the illustration are for the circulation of the heating gases and of hydrogen respectively. It is understood that coal has also been treated successfully in this plant.

It is a fact of considerable interest that the control of the process can be carried out in a building separated from the autoclave house.

Smaller plants dealing with one ton of coal per day are in operation in Germany, and H.M. Fuel Research Board have installed a similar one at Greenwich, an agreement having been entered into with Dr. Bergius and the proprietors of his patent.

The substances taking part in the reaction are supplied to the reaction vessel already heated to a temperature about 50° C. less than the reaction temperature, the coal (passing 2 mm. mesh) being made into a paste with oil and forced into the high pressure apparatus with the aid of a pump. Any hydrogen not consumed during the reaction escapes to a condensing installation for the purpose of recovery.

The following example has been given by Bergius as being typical of the yield obtained by hydrogenation of a coal (volatile matter 25 per cent.) on a comparatively large scale, namely, for about 1,000 kilogrammes.

	kgms.
Oil	435
Gas	210
Water	25
Ammonia	5
Oil and residue oil..	80
Residue	270
	1025

It is not to be wondered at that considerable interest, both from the commercial point of view and on the theoretical side should

have been taken in the Bergius process. Investigations into the feasibility of the method have proceeded in various countries since 1922, all of which it may be said have, on the whole, substantiated the claims of Bergius that a considerable proportion of the coal substance may be transformed into a tar oil by treatment at 400°–450° C. with hydrogen under high pressure. At one time considerable doubt appeared to be thrown upon the possibilities of the process by the work of Prof. Fischer and his colleagues at the Coal Research Institute, Mullheim. The majority of these latter experiments were, however, carried out without the aid of a distributing medium, and everything showed that a considerable proportion of the coal or semi-coke could be transformed into oil.

Since 1922, investigation of the Bergius process as applied to British coals, has continued without break in the Mining Research Laboratory at the University of Birmingham. These experiments (since 1924) have been financed by the British Colliery Owners' Research Association, and have, in the main, confirmed Bergius's results.

For purpose of comparison, and in order that the products of the reaction should be readily separated, it was considered advisable to suspend the coal in a dispersing medium during hydrogenation. Phenol was chosen for this purpose, because it could be readily obtained in a pure state and after the treatment in the autoclave could be completely removed by the action of caustic soda. Further, it was known to be unaffected by hydrogen at the temperature and pressure employed in the experiments. On the other hand, the employment of this material rendered the estimation of the phenols produced during hydrogenation very difficult, and in our results, quoted later, any phenols (soluble in caustic soda solution) will have been lost during the subsequent treatment of the products. Such loss explains, no doubt, to a certain extent, the average yields in our experiments being less than those obtained by Bergius. About thirty British coals have now been tested, the original coals and the products being analysed.

Hydrogenation Experiments Carried out in the Mining Research Laboratory, Birmingham University, and Financed by the British Colliery Owners' Research Association.

The samples of coal treated in these experiments (the results of which are set forth in Table II.) were ground to pass a 60 mesh I.M.M. sieve, except in two cases, where grinding was carried out in a "Colloid Circulator."

The high-pressure experiments were all carried out in a special autoclave of 3 litres capacity, shown in Plate 2. The steel of which

this was made contained 3 per cent. of nickel and 0·5 per cent. of carbon, the minimum thickness of the wall being 3 in. The apparatus is gas heated, and during the experiments is rotated at about 60 revolutions per minute in order to ensure even heating and thus minimise, as far as possible, the effects of coking on the walls of the bomb. During the progress of an experiment an asbestos-lined cover is placed over the apparatus. Temperatures are read by means of a mercury-in-steel distance thermometer which fits into a pocket penetrating through the shaft to the centre of the autoclave, from the right-hand side. The shaft on the left-hand side has a

PLATE 2.

3-litre Autoclave used in the Mining Research Laboratory Experiments.

¾-in. bore, connecting the inside and the pressure gauge, and through which gas may be admitted or discharged. The left-hand side is fitted to the body of the autoclave by means of six 1¼ in. bolts and nuts, and a pressure-tight joint is obtained by a W-shaped projection on the autoclave which fits into a thick soft-copper washer carried in the end-piece (Plate 3).

In all the experiments 200 grms. of coal were used, together with 100 grms. of phenol to act as a suspending medium. In every case it was arranged that the maximum pressure reached during the hydrogenation should be between 145 and 150 atmospheres, while the temperature was maintained, as nearly as possible, at 430° C. The total duration of heating above 400° C., in all experiments, was eight hours, and this was carried out over two days, the concen-

tration of hydrogen being maintained by blowing off the residual gases (E) after the first day's run into a gasholder, and replacing with a fresh charge of hydrogen. In every case a loss in pressure was noted at the end of each day's run.

The products of reaction were first distilled in an aluminium retort up to 150° C. The light spirit fraction (D) was separated from the water, washed with dilute caustic soda solution to remove any traces of phenol, then with dilute acid, and finally with water. The remaining products of reaction were further treated with chloroform, so that a separation into (A) phenol-insoluble, (B) phenol-soluble, chloroform insoluble, and (C) phenol-soluble, chloroform soluble was obtained. Products (B) were light brown

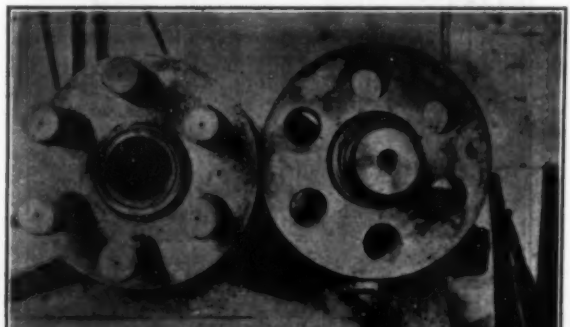

PLATE 3.
The Pressure-tight Joint Device.

powders of low apparent specific gravity, while products (C) were dark, viscous sweet smelling liquids. The products (C) were further separated into fractions distilling (1) up to 300° C., and (2) between 300° and 450° C. The yields of the various products, the results of distillation of products (C), and the iodine values, (*a*) of the light spirit fraction boiling below 150° C., and (*b*) that of the portion of product (C) distilling up to 300° C., are shown in Table II. All the iodine values were determined by Wij's method.

In one experiment, on coal from the Nottingham Top Hard Seam, to the usual charge of 200 grms. of coal and 100 grms. of pure

TABLE II.

Sample.	Destructive Distillation at 450° C. Oil yield %	Products (Ash Free Dry Basis). % Distillate to 150° C.	% Product (A).	% Product (B).	% Product (C).	Water (% on original coal).	Distillate to 300° C. % of Product (C).	Distillate to 300° C. % of original material (ash-free dry).	Distillate 300–450° C. % of Product (C).	Distillate 300–450° C. % of original material (ash-free dry).	Iodine Values. Of distillate to 150° C.	Iodine Values. Of portion of Product (C) distilling below 300° C.
Wing Vein, South Wales	9·25	—	22·6	14·5	52·3	—	25·15	13·15	19·89	10·41	—	169
No. 1 Seam, Somerset	10·80	—	18·4	18·3	33·9	—	21·40	7·25	26·30	8·92	—	291
Cockshead, N. Staffs., 60-mesh	11·90	8·30	18·20	10·30	43·65	3·88	26·16	11·42	31·76	13·86	150	179
Cockshead, N. Staffs., Colloid	—	5·35	48·91	4·71	23·09	3·53	30·45	7·03	33·02	7·62	133	169
Big Vein, South Wales	0·05	1·07	90·53	0·59	1·56	4·96	—	—	—	—	216	—
Ditto, Higher Temperature	—	2·45	87·36	0·65	2·41	4·99	—	—	—	—	138	—
High Hazels, South Yorks., 60-mesh	13·40	4·71	52·62	2·68	16·74	9·95	24·03	4·02	40·64	6·80	117	94
High Hazels, South Yorks., Colloid	—	4·65	45·56	3·14	20·82	8·83	26·81	5·58	37·62	7·83	135	145
Nine Feet, South Wales	3·55	2·12	62·94	6·62	17·86	4·73	13·75	2·45	28·97	5·16	124	143
South Fawr, South Wales	8·80	3·82	62·19	1·44	14·81	6·75	21·59	3·94	39·33	5·83	122	93
Hughes Vein, South Wales	0·20	1·62	74·33	4·66	9·72	5·34	14·96	1·45	31·87	3·10	126	358
Vitrain, Slate Seam, Warwickshire	9·05	4·28	48·76	2·58	17·35	15·91	20·97	3·64	46·57	8·08	139	309
Five Feet, Tops, Fifeshire	7·85	4·15	56·85	5·30	16·94	12·04	20·35	3·45	36·17	6·13	158	324
Namdang, India	16·40	6·32	54·08	1·21	14·38	10·68	32·80	4·72	36·33	5·22	239	186
Barnsley Softs, South Yorks.	11·65	6·34	57·99	1·05	15·77	14·67	31·92	5·03	34·17	5·39	139	144
Bullhurst, Tops, North Staffs.	10·10	6·98	24·45	9·38	38·04	8·15	27·00	10·28	31·84	12·12	148	181
Nottingham Top Hard, Soft Coal	4·60	3·98	56·80	3·63	18·15	14·80	27·92	5·07	30·96	5·62	124	120
Ditto, Catalysed with 5% Fe_2O_3	—	6·36	48·46	3·56	23·19	18·47	29·42	6·82	30·88	7·16	99	116
Nottingham Top Hard, Hard Coal	8·50	5·95	57·59	1·14	16·33	15·86	29·14	4·76	31·16	5·09	129	146
Ell Spires, Warwickshire	11·50	5·29	51·93	4·63	18·84	9·43	22·78	4·29	44·14	8·32	144	277
High Delf, Gloucestershire	10·90	5·23	40·98	5·40	31·16	9·69	25·98	8·10	31·94	9·95	137	144
Deep Seam, Kent	1·50	4·40	42·26	6·67	31·11	5·22	13·81	4·30	30·66	9·54	125	180
Ditto at lower temperature	—	2·22	53·81	6·57	23·05	4·80	12·60	2·90	28·69	6·61	209	253
South Staffs. Thick Coal, White Seam												
Clarain	—	—	21·8	6·0	37·7	—	—	—	—	—	—	—
Durain	8·10	—	37·5	7·5	37·5	—	—	—	—	—	—	—
Graigola	—	—	26·5	33·1	26·1	—	—	—	—	—	—	—
Arley Seam, Lancs.	—	—	16·4	6·2	61·4	—	—	—	—	—	—	—
Brown Coal, Cologne	11·30	—	24·9	17·4	39·7	—	14·4	5·72	—	—	—	—
Lignite, Devon	5·25	—	20·3	20·3	40·3	—	38·5	15·52	—	—	—	—
Lignite Semi-coke, Devon	—	—	59·6	10·9	24·2	—	24·0	5·81	—	—	—	—
Recent Lignite, Devon	18·75	—	16·3	3·6	29·3	—	28·2	8·3	—	—	—	—
Semi-coke from recent Lignite, Devon	—	—	79·3	2·9	6·1	—	18·6	1·13	—	—	—	—
Boghead Cannel, Fifeshire	35·40	3·21	31·9	nil	48·4	3·35	38·64	19·90	37·98	19·44	675	—
Cannel Coal, South Staffs., Thick Coal	21·00	6·82	46·3	1·9	28·1	—	24·30	8·48	34·54	12·04	226*	—

* Whole distillate to 300° C.

TABLE III.

Sample.	Proximate %. Moisture.	Ash.	Volatile (less moisture)	Fixed Carbon.	Ultimate % (Ash Free Dry Basis). Carbon.	Hydrogen	Nitrogen.	Sulphur.	Oxygen.	C/H Ratio.
Wing Vein, South Wales	0·85	3·41	32·61	63·13	85·77	5·11	1·59	0·79	6·74	16·75
No. 1 Seam, Somerset	1·74	3·65	34·36	39·75	81·71	5·32	1·49	1·54	9·94	15·36
Cockshead, North Staffs.	1·06	2·65	34·38	61·91	85·81	5·22	1·95	0·54	6·48	16·44
Big Vein, South Wales	2·03	4·18	7·95	85·84	92·64	3·30	0·82	0·89	2·35	28·06
High Hazels, South Yorks.	3·98	2·16	39·41	54·45	82·86	5·10	1·22	1·01	9·81	16·25
Nine Feet, South Wales	1·68	2·81	17·26	78·25	87·80	4·63	1·28	0·85	5·44	18·96
South Fawr, South Wales	2·92	4·06	28·24	64·78	86·16	5·24	1·27	1·15	6·18	16·44
Hughes Vein, South Wales	3·07	3·61	11·64	81·68	91·43	3·87	1·10	1·00	2·60	23·62
Vitrain, Slate Seam, Warwickshire	8·75	2·16	36·61	52·48	79·83	5·74	1·29	1·11	12·03	13·91
Five Feet, Tops, Fifeshire	7·42	3·93	27·74	60·91	85·91	4·83	1·22	0·87	7·17	17·79
Namdang, India	3·98	0·55	39·20	56·27	83·08	5·42	1·22	1·17	9·11	15·33
Barnsley Softs, South Yorks.	8·40	2·46	34·05	55·09	83·05	5·23	1·72	0·83	9·17	15·88
Bullhurst, Tops, North Staffs.	2·05	4·43	33·10	60·02	84·03	5·48	1·35	2·49	6·65	15·33
Nottingham Top Hard, Soft Coal	11·53	4·89	31·22	52·36	81·55	4·76	1·76	1·61	10·32	17·13
Nottingham Top Hard, Hard Coal	10·15	5·62	30·13	54·10	83·08	4·74	1·54	0·94	9·70	17·53
Ell Spires, Warwickshire	5·22	9·70	38·7	46·4	80·16	5·76	1·00	1·36	11·72	13·92
High Delf, Gloucestershire	4·33	4·43	35·27	55·97	83·77	5·56	1·10	1·36	8·21	15·07
Deep Seam, Kent	1·71	6·85	20·63	70·81	89·66	4·60	1·36	1·84	2·54	19·49
South Staffs., Thick Coal, White Seam										
Clarain	13·26	1·44	33·37	51·93	81·25	5·27	1·25	0·94	11·29	15·41
Durain	3·50	6·06	31·18	59·26	82·44	5·08	1·04	0·72	10·72	16·23
Graigola	1·36	6·76	13·28	78·60	91·54	4·32	1·45	0·84	1·85	21·19
Arley Seam, Lancs.	1·92	2·87	33·68	61·53	84·19	5·47	1·86	1·79	6·69	15·39
Brown Coal, Cologne	16·99	2·52	44·47	36·02	69·01	4·93	0·66	0·89	24·51	14·00
Lignite, Devon	32·38	21·86	38·90	6·86	65·69	5·53	0·99	2·11	25·68	11·88
Lignite, Semi-coke, Devon	0·64	44·90	23·80	30·66	79·58	4·01	1·32	1·89	13·20	19·85
Recent Lignite, Devon	20·06	1·16	49·30	29·48	69·63	6·16	0·22	1·26	22·73	11·30
Semi-coke from recent lignite, Devon	1·01	4·01	41·80	53·18	80·42	4·56	0·28	1·11	13·63	17·64
Boghead Cannel, Fifeshire	1·07	25·82	55·58	17·53	78·50	9·81	1·05	4·04	6·60	8·00
Cannel Coal, South Staffs., Thick Coal	5·20	3·35	46·07	45·38	78·63	6·47	1·52	1·69	11·69	12·1

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phenol, was added 8·4 grms. of ferric oxide to act as a catalyst, this representing 5 per cent. of the ash-free dry coal. The subsequent separation was carried out in the usual manner.

TABLE IV.

Sample.	Petrol yield (1) Galls. per ton of original coal.	True tar yield (2) (Product C). Galls. per ton of original coal.	Total oil (1) + (2). Galls. per ton of original coal.
Wing Vein, South Wales	—	102	—
No. 1 Seam, Somerset	—	65	—
Cockshead, North Staffs.	23·0	86	109·0
Ditto, ground in colloid mill	14·5	45	59·5
Big Vein, South Wales	2·8	3·0	5·8
Ditto, at higher temperature	6·4	4·6	11
High Hazels, South Yorks.	12·5	33	45·5
Ditto, ground in colloid mill	12·5	41	53·5
Nine Feet, South Wales	5·5	35	40·5
South Fawr, South Wales	10·0	28	38
Hughes Vein, South Wales	4·3	18·5	22·8
Vitrain, Slate Seam, Warwickshire ..	11	31·5	42·5
Five Feet, Tops, Fifeshire	10·5	31	41·5
Namdang, India	17	28	45
Barnsley, Softs, South Yorks.	16	28·5	44·4
Bullhurst, Tops, North Staffs.	18·5	72·5	91
Nottingham Top Hard, Soft Coal	9·5	31	40·5
Ditto, catalysed with 5% Fe_2O_3	15	39·5	54·5
Nottingham Top Hard, Hard Coal ..	14	28	42
Ell Spires, Warwickshire	12·5	32·5	45
High Delf, Gloucestershire	13·5	58	71·5
Deep Seam, Kent	11·5	58	69·5
Ditto, at lower temperature	5·7	42·5	48·2
South Staffs. Thick Coal, White Seam. ..			
Clarain	—	65·5	—
Durain	—	69·5	—
Graigola, South Wales	—	49	—
Arley, Lancs.	—	119	—
Brown Coal, Cologne	—	65	—
Lignite, Devon	—	38	—
Lignite Semi-coke, Devon	—	27	—
Recent Lignite, Devon	—	47	—
Recent Lignite Semi-coke, Devon	—	11·8	—
Boghead Cannel, Fifeshire	6·5	72	78·5
Cannel Coal, South Staffs., Thick coal ..	17·5	52·5	70·0

These figures are obtained by taking the average figure 0·8 as the density for the petrol fraction, and that of 1·1 for Product (C).

On reference to Table II. it will be seen that the yield of petrol was, proportionally, most increased, while a somewhat larger amount of tar-oil was also produced. There was a substantial increase in the amount of aqueous liquor, but the yield of product (B) was practically unaffected. Further experiments on the effect of catalysts are being carried out.

In one column of Table II. there is given for comparison the yield of tar-oil obtained by destructive distillation at 450° C.

The proximate and ultimate analyses of the coals are given in Table III.

Table II. shows that, in general, the yields of tar-oil (product (C)) are less than those obtained by Bergius. This is no doubt partly due to the production of phenols, which, as already explained, will appear as "loss" in our tests owing to the method adopted for treatment of the products. Moreover, the pressures employed by us are, on the whole, less than those used by Bergius in small scale work, although of about the same order as that pressure employed in his large size plant. The effect of raising the hydrogen pressure is shown by our experiment with coal from No. 1 Seam, Somerset. The yield of tar-oil has been increased by approximately 6 per cent. of the original coal. We consider, therefore, that under slightly higher pressures yields greater than those shown will probably be obtained. Table IV. gives the yield of petrol and tar-oil calculated in gallons per ton of coal (as received), taking the specific gravities of the two at 0·8 and 1·1 respectively.

From these results it will be clear that under the same working conditions some coals are much more amenable than others to the action of hydrogen under high pressure. Our aim has been, and is, to ascertain with more exactitude than appears to have been the case with the experiments carried out by the Bergin Actien Gesellschaft, which portions of the coal substance undergo hydrogenation most readily. Product (B), the amount of which, it will be noted, varies with different coals, is undoubtedly a product of hydrogenation. Further research into the nature of this portion will, we believe, afford useful evidence regarding the course of the hydrogenation reaction.

Reference has been made to Bergius's view that certain coals are converted to oil in greater amount than others. In his early patents and writings he emphasised the point that coals containing more than 85 per cent. of carbon on an ash-free dry basis were difficult to hydrogenate. Although generally true, there are undoubtedly certain types of coal which appear to fall outside the scope of this statement. Undoubtedly considerably more research is desirable before the method can be considered to be more than a purely empirical one.

Products.

Tables I and II indicate that the solid residue left after hydrogenation will amount, on the average, to about 20 per cent. of the original coal, and will contain all the ash of the latter. The com-

mercial utilisation of this product seems difficult except in the form of powdered fuel or in low grade producer gas practice.

The *oil* is, of course, the principal product of the reaction. Bergius considers that its composition is quite different from that for either high or low temperature carbonisation. Instructive information has been given by Ormandy and Craven (*J. Inst. Petroleum Technol.* 12 (1926) pp. 77-88), regarding the composition of the petrol or "gasoline" fractions obtained by the hydrogenation of a South Yorkshire coal (Orgreave washed slack). The petrol amounted in yield to approximately 10 per cent. of the coal, and consisted, as will be seen from the figures given below, mainly of paraffin and naphthene hydrocarbons.

	%
Unsaturated hydrocarbons	3·1
Aromatic hydrocarbons—benzene, toluene, xylene	47·0
Paraffins—Butanes, pentanes, hexanes	47·0
Naphthenes—Cyclopentanes and cyclohexanes ..	27·0
Residue	15·4
	100·0

Nearly 85 per cent. of this petrol fraction would thus appear to consist of hydrocarbons. On the other hand, tests carried out in this laboratory indicated that the kerosine fraction* (boiling below 300° C.) contained a considerable proportion of oxygenated bodies. This fraction gave 9 per cent. of oxygen on analysis.

The yield of gaseous products in hydrogenation is as great as that obtained in normal gas works practice, forming about 20 per cent. by weight of the original coal. It seems, therefore, that a considerable proportion of the carbon of the coal is split off in the form of methane and ethane as the result of interaction with hydrogen.

The results given in Table II indicate that the petrol and kerosine fractions of the hydrogenation product (in the Birmingham tests) are still unsaturated, but would, by no means, be objectionable in this respect for utilisation in internal combustion engines.

General Conclusions and Discussion from the Economic Aspect.

It has been made evident by the results quoted that the treatment of coal by hydrogen under high pressure gives a yield of oil far in excess of that obtained by any other method of coal treatment, although we feel that the original patent claims of Bergius and

* From the hydrogenation of vitrain from Warwickshire slate coal.

his colleagues were somewhat more optimistic than subsequent tests have been able to justify.

The nature of the oily products, is of course of prime importance from the financial aspect.

The results obtained by Ormandy and Craven, as well as our own, carried out on a smaller scale, confirm the view that petrol suitable for internal combustion engines may be obtained in amount up to 25 gallons per ton of coal treated. The value of the heavier oil is not, however, so assured. Bergius claims that good lubricating oils have now been obtained from the crude heavy oil, and that the phenolic portion contains simple phenols, cresols and xylols without the objectionable phenols of higher molecular weight. The market value of the third main product, namely, the gas, in spite of its high calorific value, will probably not be great unless the process can be worked in conjunction with a gas-making plant.

So far as one can see at the moment it would appear that the value of the products will mainly depend upon the prevailing price of petrol and fuel oil.

Assuming the price of petrol and fuel oil to be maintained at their present selling value, and taking a figure of 15 per cent. motor spirit together with 45 per cent. of fuel oil, the retail value of the products should be approximately £4 per ton of coal treated, calculated as follows:—

	s.	d.
0·15 tons motor spirit at 1s. per gallon	42	0
0·45 tons fuel oil at £4 per ton	36	0
	78	0

Coming now to the costs of the process, 30s. would seem, at the present time, to be about the figure for the amount of hydrogen necessary, and 15s. for the cost of coal. A margin of 33s. will be left to cover all other costs of working, labour, depreciation of and repairs to plant, and interest on capital outlay of plant. The latter will obviously be very high, as will also, no doubt, the depreciation and repair items. It is quite certain, however, that the prime necessity for the commercial success of the process is cheap hydrogen. The crude calculation given above has been based upon the cost of production of hydrogen by the steam-iron reaction using either "blue" water gas, or cheap coke oven gas.

Bergius, of course, realises this necessity for cheap hydrogen, and has, in consequence, developed a new process in which the gaseous products of the hydrogenation are utilised. This process is based upon the interaction of steam and methane.

Attention has already been drawn to the fact that certain coals have now been proved to be more suitable than others for this method of treatment. Although one cannot limit the most suitable coals to a definite group in any of the present methods of classifying coal, we can say from the researches carried out in the Mining Research Laboratory that, of the coals so far examined, those falling into the parabituminous division of the Seyler Classification and having a carbon/hydrogen ratio between 15·5 and 16·5 appear to give the highest oil yields.

Bergius has stated that, with certain types of lignite, he has obtained almost complete conversion of the coal to oil and gas. This has not been our experience, and no doubt the difference is due to fundamental differences in the chemical constitution of the coals tested.

With coals of the class referred to above, the process could possibly be worked at the present time with profit. The annual output of such coals is sufficient to supply the greater bulk of the present day fuel oil requirements of this country. There is no doubt in our minds, however, that it is only by further researches into the influence of constitution of coal upon ease of hydrogenation, and the nature of the products obtained as well as further research in the engineering technique of high-pressure work, that the more general application of the process can be made a commercial success.

In conclusion, we wish to thank the British Colliery Owners' Research Association for permission to publish the results included in this paper, and also Prof. J. S. Haldane, Director of this Laboratory and President of the Institution of Mining Engineers, for his interest and advice, always so helpful in whatever line of research it is given.

The Effect of Thermometric Lag in Fractional Distillation.*

W. R. Ormandy, D.Sc., F.I.C., F.C.S., M.I.A.E., and
E. C. Craven, B.Sc., A.Inst.P., F.C.S.

The operation of fractional distillation is so frequently carried out in chemical laboratories that it is of importance to inquire if any serious source of error has been overlooked therein. Such a distillation may have for its object the determination of the degree of purity of some body, or the recognition and separation of the components of a mixture.

Some time ago the authors made an extended investigation[1] into the effect of the lag of mercurial thermometers in distillations carried out from ordinary distilling flasks of the Engler type. The magnitude of the effect found was surprising. For example, with a petroleum spirit boiling roughly from 60° C. to 180° C. the means of the lags of six thermometers were as follows :—

	° C.
Drop A (1st drop from side tube of flask)	15·7
B (,, condenser)	12·4
10% distilled	3·7
30% ,, ...	2·2
50% ,, ...	0·8
70% ,, ...	1·9
90% ,, ...	2·2
Dry point	6·8

The lags were determined by carrying out duplicate distillation, using a thermo-couple to determine the vapour-temperatures. The couple was of bare 26-gauge wires and was used with the junction pointing upwards so that liquid condensing elsewhere could not run down over the junction. It was shown that the lag of such a junction is entirely inappreciable for the present purpose.

In dealing with pure liquids it was found that the reading of the mercurial thermometer rose steadily over the whole course of the distillation, whilst the thermo-couple reading might remain constant from drop to dry point. Even at the close of the distillation there was usually a distinct outstanding lag of the thermometer.

The present work follows along similar lines, but an 8-bulb Young "Evaporator" still head was used throughout. The results here given are confined to the I.P.T. low "Distillation" thermometer No. 243133 mentioned in the previous Paper, but a few trials

* Paper received December 4th, 1927.
[1] *J. Inst. Petr. Techn.*, 1024, **10**, 342.

with other thermometers have shown that the lag effects are very much of the same order. The reason for this will be dealt with shortly.

When a cold thermometer is plunged into the vapour of a boiling liquid itself it has been found that the initial consequent rapid rate of rise of the thermometer reading closely follows Newton's law :—

$$d\theta/dt = k(\theta_{max} - \theta) = k\phi$$

where θ = reading after time t.

θ_{max} = reading after time $t = \infty$

ϕ = lag.

The constant k may be termed the " speed " of the thermometer and represents the number of degrees the thermometer would rise per second if the bath were maintained always 1° above the thermometer reading.

The speed k for different thermometers depends on the diameter of the bulb and the thickness of its walls. Comparative measures indicate that for cylindrical bulbs the length is not of importance. For the same thermometer in a vapour, k depends on the latent heat of condensation of the vapour and the speed at which it passes the bulb. In a liquid bath k appears to depend on the rate of stirring.

The speed constant may be determined by plunging the cold thermometer into the vapour or liquid and determining the rate of rise of the mercury.

$$k = 2{\cdot}30\ (\log_{10}\phi_1 - \log_{10}\phi_2)\ (t_2 - t_1)$$

The following results have been obtained on the thermometer No. 243133 previously mentioned :—

	k.
In stirred oil bath	0·23
In tube of mercury heated by boiling water ..	0·33
In boiling water	0·46
" stirred	0·67
" stirred faster	0·76
*In air	0·07
In heptane vapour 1 drop per sec.	0·26
" 2 drops per sec.	0·30
In slow steam	0·29
In steam distilling 2 drops per sec.	0·58

* By cooling curve in air.

It may be of interest to repeat the following table for a very similar thermometer for value of k determined in vapours at identical rates of distillation :—

Liquid.	k.	Latent ht. cals./gm.
Heptane	0·41 ..	76
Alcohol	0·50 ..	207
Water	0·72 ..	539

It will be noted the variations in k follow the rules already given, and it may be noted that in general a thermometer rises quite as rapidly in the vapour from a boiling liquid as in the liquid itself. In distillations from an Engler flask the liquid at 2 drops per second is distilling at the rate of 3 c.c. per minute. The area of the neck of the flask is about $2\frac{1}{2}$ sq. cms. The vapour speed is therefore about 20 metres per minute or 1 ft. per second. In the experiments with the fractionating column the usual 2 drops per second rate was adhered to, since at slower rates the effect of draughts is rather pronounced.

It would appear in general that the initial lag of the mercurial thermometer is dependent on the thickness of the layer of condensed liquid on the bulb, or in the case of a liquid bath on a layer of cooled liquid around the bulb. Consider a section of the bulb when in operation in vapour.

Let r = radius of mercury.
g = thickness of glass.
l = thickness of liquid layer.
K_g, K_l = thermal conductivities of glass and liquid respectively.

Assume the thermometer to be rising steadily so that a constant difference $\theta_m - \theta$ is maintained between the vapour temperature and the mercury temperature. Owing to the relatively high thermal conductivity of mercury the latter may be taken to be at an even temperature throughout.

Let θ' = temperature of liquid—glass surface.

Then heat crossing any isothermal per sec. per cm. length.

$$= H = 2\pi K_g(\theta' - \theta)/\log(r+g) - \log r \doteqdot 2\pi r K_g(\theta' - \theta)/g$$
$$= 2\pi K_e(\theta_m - \theta')/\log(r+g+l) - \log(r+g) \doteqdot 2\pi r K_l(\theta_m - \theta')/l$$

Also $H = C\, d\theta/dt = kC(\theta_m - \theta)$

where C = heat capacity of the mercury.

Eliminating θ'

$$l = K_l(2\pi r/kC - g/K_g)$$

The thermal conductivities are roughly as follows:—

Water	0·0014
Heptane	0·0006
Glass	0·0025

For thermometer 243133

$$r = 0{\cdot}325 \text{ cms.}$$
$$g = 0{\cdot}015 \text{ cms.}$$

These were determined by immersing the thermometer in a liquid whose refractive index was adjusted to be nearly as possible that

of the glass of the thermometer bulb. The glass thickness was observed to be very uneven but a rough mean value is given

$$C = 0{\cdot}15 \text{ cals/cm.}$$

The values of k under various conditions have already been given.

From these data the thickness l of the liquid film has been computed to be

	l cms.
In heptane 1 drop per sec.	0·025
„ 2 drops per sec.	0·022
water (slow steam)	0·052
„ 2 drops per second	0·022

The thickness of the liquid film required to produce the observed lags is therefore of the order of a quarter of a millimetre.

It may be noted that if no liquid film were formed the speed factor k of the thermometer would be $2\pi K_g r/gC = 2{\cdot}2$ i.e. the thermometer speed would be from four to ten times greater than is actually observed.

In the previous paper it was stated that it was possible the last slow rise of the thermometer is dependent more on the heating of the stem, and consequent expansion of the mercury thread, than on the passage of heat into the bulb. For the last few tenths of a degree rise, therefore, it was thought the logarithmic law might not hold exactly. This idea has been verified in the present work, and whilst the correction is not important in the case of the distillation of mixtures with a wide boiling point range it is of some importance in the case of pure liquids. Experiments have shown that the total lag of a mercury thermometer may be expressed as the sum of two terms representing the lag due to the bulb and the stem respectively. Thus if

ϕ = Total lag at time t
ϕ_B = Initial lag due to bulb.
ϕ_S = Initial lag due to stem

we have

$$\phi = \phi_B \epsilon^{-k_1 t} + \phi_S \epsilon^{-k_2 t}$$

The first term becomes negligibly small after 1 minute, but the second term is evident until after 15 or 20 minutes, i.e. till 50 c.c. or so has been distilled. For heptane distilled with the column the values were found to be

$$\phi = 77{\cdot}04\, \epsilon^{-{\cdot}279\, t} + {\cdot}86\, \epsilon^{-{\cdot}00353\, t}$$

The observed and calculated lags are shown in the graph.

The quantity ϕ_s must not be confused with the ordinary corrections for emergent stem. The latter gives the increase which would be obtained if the whole thread were heated to the

temperature of the bulb whilst ϕ_s is the increase which would be observed if the exposed thread were heated from room temperature to its final mean temperature. The latter could no doubt be calculated by applying the usual linear heat conduction equation in which case the thermal conductivity and emissivity of the stem would have to be determined.

It would appear from the foregoing that the lags observed are due, therefore, chiefly to the liquid film about the bulb of the thermometer in operation, and to the lag in attainment of stem temperature equilibrium. This explains why the observed lags do not differ greatly from one thermometer to another.

In the following distillations using the 8-section column, the distillation speed was kept as closely as possible to 2 drops per second. The thermometer was calibrated in an oil bath against the couple, the latter being taken as correct. The depth of immersion was made as nearly as possible that which would obtain in the distillation. A correction was required to the calibration curve obtained in this way owing to the fact that the air temperature above the oil bath is greater than the air temperature above the column, for any given temperature of the latter and the oil bath. This correction was determined at several points by carrying out the distillation of larger quantities of pure liquids through the column until the thermometer was steady. The outstanding lags at the close of distillation referred to in the previous paper were due to the neglect of this correction.

In most cases the couple and thermometer were inserted together into the neck of the distilling column, the junction, however, being set well clear of the bulb. The volume of liquid used in each case was 100 c.c. Owing to the "hold-up" of the column the last 10-15 per cent. could not be distilled over. Where the temperature was not rising rapidly the readings are believed to be correct to 0·1° C.

PURE LIQUIDS. METHYL ALCOHOL.

	Couple. °C.	Thermo. °C.	Lag. °C.	Lag Flask. °C.
Drop A	64·3	63·0	1·3	0·8
B	64·3	64·0	0·3	0·7
1% over	64·3	64·0	0·3	0·5
2% „	64·3	64·0	0·3	0·5
5% „	64·3	64·1	0·2	0·2
10% „	64·3	64·2	0·1	0·1
20% upwards ..	64·3	64·3	Nil.	0·1

The last column refers to the lags determined in distillations from an Engler flask with a very similar thermometer. The figures are given for comparison. In the particular case of methyl

alcohol, however, that used in the flask distillation was much less nearly pure.

BENZENE.

	Couple. °C.	Thermo. °C.	Lag. °C.	Lag Flask. °C.
Drop A	79·0	70·0	9·0	4·1
B	79·9	79·0	0·9	1·0
1% over	79·9	79·4	0·5	1·0
2% „	79·9	79·5	0·4	0·9
5% „	79·9	79·6	0·3	0·7
10% „	80·1	79·7	0·4	0·6
20% „	80·1	79·8	0·3	0·5
30% „	80·1	79·9	0·2	0·4
40% „	80·1	80·0	0·1	0·3
50% „	80·1	80·1	Nil	0·3
60% „	80·1	80·1	Nil	0·2
70% „	80·1	80·1	Nil	0·2
80% „	80·1	80·1	Nil	0·2
90% „	80·1	80·1	Nil	0·1

With benzene, in spite of careful drying, drop A was always turbid with water.

HEPTANE.

	Couple. °C.	Thermo. °C.	Lag. °C.	Lag Flask. °C.
Drop A	97·9	91·5	6·4	1·8
B	97·9	96·8	1·1	0·9
1% over	98·1	97·6	0·5	0·7
2% „	98·1	97·6	0·5	0·7
5% „	98·1	97·6	0·5	0·7
10% „	98·1	97·7	0·4	0·7
20% „	98·1	97·8	0·3	0·6
30% „	98·1	97·9	0·2	0·4
40% „	98·1	98·0	0·1	0·3
50% „	98·1	98·0	0·1	0·3
60% „	98·1	98·1	Nil	0·2
70% „	98·1	98·1	Nil	0·2
80% „	98·1	98·1	Nil	0·2
85% „	98·1	98·1	Nil	

TOLUENE.

	Couple. °C.	Thermo. °C.	Lag. °C.	Lag Flask. °C.
Drop A	109·8	99·1	10·7	2·3
B	110·4	108·8	1·6	1·1
1% over	110·5	109·6	0·9	0·9
2% „	110·5	109·7	0·8	0·5
5% „	110·5	109·7	0·8	0·4
10% „	110·5	109·8	0·7	0·4
20% „	110·5	110·0	0·5	0·4
30% „	110·5	110·2	0·3	
40% „	110·5	110·3	0·2	
50% „	110·5	110·4	0·1	
60% „	110·5	110·4	0·1	
70% „	110·5	110·4	0·1	
80% „	110·5	110·5	Nil	
85% „	110·5	110·5	Nil	

Mixtures. Ether-isopropyl Alcohol.	Couple. °C.	Thermo. °C.	Lag. °C.
8% over	34·2	33·9	0·3
9% „	34·6	34·2	0·4
10% „	34·9	34·7	0·2
11% „	41·8	38·0	3·8
12% „	78·8	75·8	3·0
13% „	80·0	79·8	0·2
14% „	80·2	80·0	0·2
15% „	80·2	80·1	0·1
16% „	80·2	80·2	Nil

The mixture benzene-toluene is classic in fractionation experiments. A 1 vol.: 3 vols. mixture gave the following figures:—

	Couple. °C.	Thermo. °C.	Lag. °C.
8% over	84·8	83·7	1·1
10% „	85·2	84·1	1·1
12% „	85·7	84·5	1·2
14% „	86·0	84·9	1·1
16% „	86·4	85·1	1·3
18% „	86·9	85·4	1·5
20% „	87·8	86·2	1·6
22% „	89·6	88·4	1·2
24% „	91·7	90·3	1·4
26% „	94·3	92·9	1·4
28% „	97·0	95·7	1·3
30% „	99·5	98·2	1·3
32% „	102·1	101·0	1·1
34% „	104·8	104·2	0·6
36% „	107·2	106·4	0·8
38% „	108·4	107·7	0·7
40% „	108·8	108·3	0·5
42% „	109·0	108·7	0·3
44% „	109·3	108·9	0·4
46% „	109·3	109·2	0·1

Petroleum Spirit (1).

	Couple. °C.	Thermo. °C.	Lag. °C.
Drop A	35·8	31·9	3·9
B	38·7	36·1	2·6
1% over	57·5	55·7	1·8
2% „	65·0	63·2	1·8
5% „	73·2	71·5	1·7
10% „	80·9	79·4	1·5
15% „	87·9	86·7	1·2
20% „	93·6	92·2	1·4
25% „	99·6	98·5	1·1
30% „	102·9	102·2	0·7
35% „	106·0	105·3	0·7
40% „	108·0	107·3	0·7
45% „	112·0	111·3	0·7
50% „	116·4	115·7	0·7

Petroleum Spirit (2) The following portion of a distillation is of particular interest :—

	Couple. °C.	Thermo. °C.	Lag. °C.
26% over	73·8	73·1	0·7
28% „	74·7	74·0	0·7
30% „	75·6	75·0	0·6
32% „	75·9	75·7	0·2
34% „	76·9	76·3	0·6
36% „	77·4	76·9	0·5
38% „	77·7	77·4	0·3
40% „	78·1	77·9	0·2
42% „	78·8	78·4	0·4
44% „	79·6	79·2	0·4
46% „	80·9	80·5	0·4
48% „	81·4	81·1	0·3
50% „	83·1	82·8	0·3

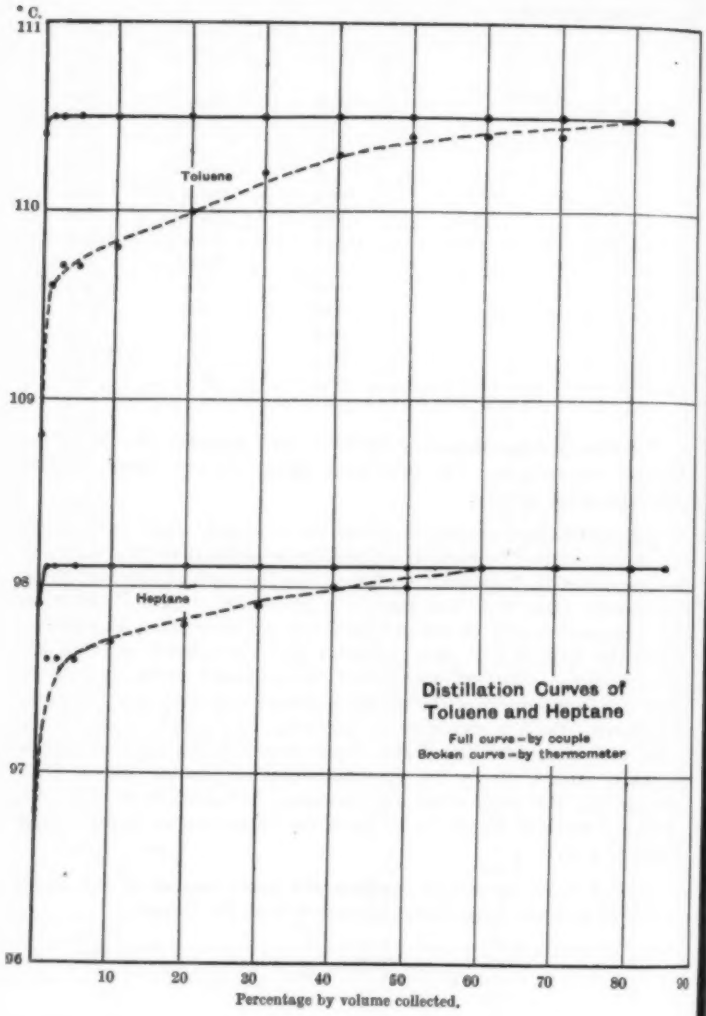
The distillation curves of heptane and toluene, the benzene-toluene mixture and the petroleum spirit (2) are shown in the accompanying graphs.

The distillation examples given show clearly that very great caution must be observed in accepting the indications of a mercury thermometer in fractionation tests. In the case of liquids which are nearly pure the slow rise of temperature during distillation must not necessarily be taken to indicate the presence of impurities. Since the bulk of this slow rise after the first minute or so is due to conduction of heat along the thermometer stem, it follows that total immersion of the mercury thread would greatly minimise this error. Such a procedure is, however, in general, incompatible with the usual position of the thermometer bulb. The best plan perhaps would be to use the thermometer upside down, thus minimising the stem error and drainage of liquid over the bulb. Another method would be to have the thermometer made in the form of a U.

It is of some interest to compare the lags observed in the distillation of heptane with those calculated from the formula

$$\phi = \phi_B \epsilon^{-k_1 t} + \phi_S \epsilon^{-k_2 t}$$

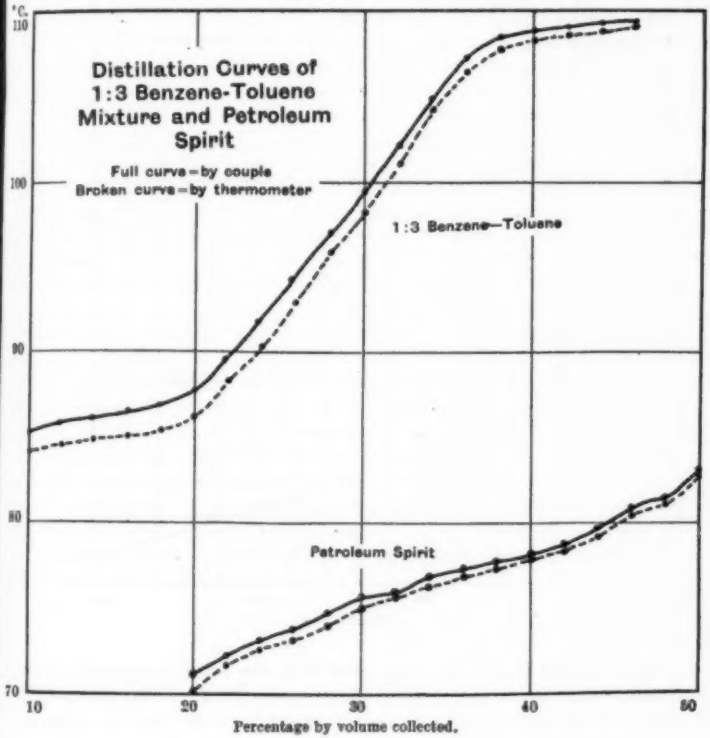
The time t from the first contact of the vapour with the bulb to the various stages of the distillation may be taken as in the corres-


°C.
111
110
109
98
97
96
Toluene
Heptane
Distillation Curves of
Toluene and Heptane
Full curve – by couple
Broken curve – by thermometer
10
20
30
40
50
60
70
80
90
Percentage by volume collected.

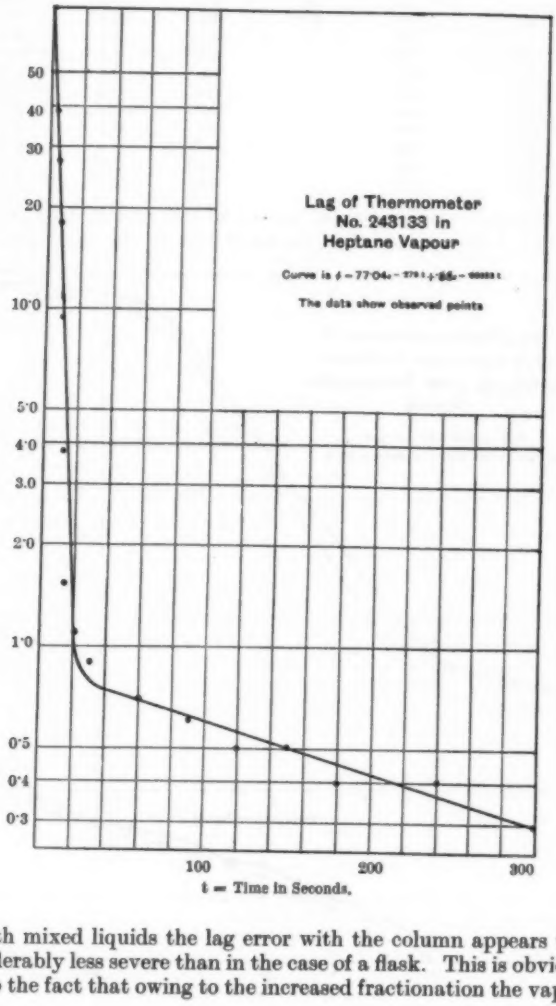
ponding distillation from a flask as previously determined. The following results are obtained for heptane :—

	t. Secs.	ϕ B.	ϕ S.	ϕ Calc.	ϕ observed.
Drop A ..	12	3·2	0·8	4·0	6·4
B ..	28	0·04	0·8	0·8	1·1
1% ..	49	—	0·7	0·7	0·5
2% ..	70	—	0·7	0·7	0·5
5% ..	134	—	0·6	0·6	0·5
10% ..	240	—	0·4	0·4	0·4
20% ..	400	—	0·2	0·2	0·3
30% ..	560	—	0·1	0·1	0·2
40% ..	720	—	0·1	0·1	0·1
50% ..	888	—	0·04	Nil	0·1
60% ..	1040	—	0·02	Nil	Nil

The differences are not great except in the case of drop A. So frequently, however, are the calculated lags in the early parts of



distillations found to be lower than those observed that it is difficult to resist the conclusion that the cold neck of the flask or column has the effect of increasing the initial lags.



With mixed liquids the lag error with the column appears to be considerably less severe than in the case of a flask. This is obviously due to the fact that owing to the increased fractionation the vapours

around the thermometer will be more nearly of uniform volatility. The examples given show, however, that at turning points in distillations, considerable lags do occur, and that the damping effect of the thermometer may be sufficient to obscure the indications of small amounts of components in the mixture. If an attempt be made to calculate the lags to be expected in distillations of mixtures the difficulty arises that the distillation curve cannot be expressed as a known function of the time. This was overcome in the previous paper by dealing with the curve in small sections, say 10-second time intervals, when the equation may be written :—

$$d\theta/dt = k(a + bt - \theta)$$

where a=temperature of vapour at start of time interval and bt=temperature rise over the interval. The solution of this equation led to the result that the lags should be $b/k - (\theta_0 + b/k - a)\epsilon^{-kt}$ where θ_0 is the thermometer reading at the start of the time interval. The term b/k is the lag arising from the rate of temperature rise of the vapour, the remainder of the expression is the lag arising from the initial temperature of the thermometer. In point of fact the lags so calculated were far below those observed. For example, with a petroleum spirit the lag observed after 200 seconds was 4·0° C. whilst the calculated lag was a small fraction of a degree. Even if a correction be applied for exposed mercury thread the discrepancy would be but little affected. It would appear that the temperature of the vapour surrounding the thermometer bulb is greatly reduced by the condensation of the higher boiling point fractions of the mixture on the bulb, so that the temperature head is less than is assumed in the calculations. This effect must also occur to some extent with a thermo couple, but probably only to a negligible extent.

The subject hardly merits much further treatment. Sufficient evidence has been given to show that vapour temperatures as measured by a mercury thermometer are always liable to be in error, and that in distillations of any importance, the use of a couple is greatly to be preferred.

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Considerations on the Vapour-Phase Oxidation of Aromatic Hydrocarbons and of Petroleum Distillates.*

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The subject of the possible treatment of petroleum as a chemical raw material is constantly referred to in the more popular scientific press, and analogies are drawn with the use of coal tar for the production of intermediates. This possibility has been well reviewed by B. T. Brooks[1]. This review, written as it is by an authority, shows clearly the difficulties which beset any immediate commercial application and that the slow development of petroleum chemistry is due to our deficient knowledge of the higher aliphatic hydrocarbons and their isolation. The possibility of the conversion of certain petroleum distillates by vapour phase oxidation is without doubt a promising mode of attack. The problem so far as petroleum is concerned divides itself into two main branches of study; firstly the investigation of the large number of claims for successful oxidation of many pure aromatic hydrocarbons, and secondly their application to petroleum distillates. The latter includes a knowledge of the isolation and purity of readily obtainable components of petroleum necessary as starting materials. The history of the development of vapour phase oxidation processes and earlier work will also be of great value, as much of the more recent work is mostly concerned with such aromatic hydrocarbons as naphthalene and anthracene.

Coquillion[2] was one of the first to study the partial oxidation of substances by the passage of suitable mixtures of substances with air over incandescent platinum and palladium wire. In this way toluene was found to form bitter almond oil and later[3] benzene, xylene, and cumene, were also studied. Walter[4] used heated copper spirals for the partial oxidation of air-alcohol mixtures, and also, was the first to employ the effective partial oxidation catalyst, vanadium pentoxide. Using a catalyst made by the ignition of asbestos soaked in ammonium vanadate he obtained benzoic acid

* Paper received December 9th, 1927.
[1] *J. Ind. Eng. Chem.*, 1924, **16**, 185.
[2] *Compt. rend.*, **77**, 444.
[3] *Compt. rend.*, **80**, 1089.
[4] *J. prakt. Chem.*, 1895 (2), **51**, 107.

and benzaldehyde (5·7%) from air-toluene mixtures. In his later patent,[5] besides the formation of formaldehyde from methyl alcohol, he gives the following as "instance III." A mineral oil distillate boiling above 300° C. is sprayed into a current of air mixed with steam and the mixture is led over asbestos coated with finely divided copper oxide (at red heat). The latter keeps its heat during the reaction. The products obtained include acetic acid and its homologues, aldehydes (solid), organic acids (solid at 0° C.), and oils, of which a half distilled between 140–300°. The oxidation of alcohol-air mixtures was studied by Trillat.[6] The varied nature of the products that can be derived from a pure hydrocarbon is well shown by the work of Stepski,[7] who led various vapour-air mixtures over glowing platinum and then through a condenser at − 15°, while gaseous olefines were trapped in bromine water. In this way iso-pentane gave formaldehyde, ethylene, propylene, $\Delta^{\alpha\beta}$ and $\Delta^{\beta\gamma}$ butylene, iso-butylene, two iso-amylenes, butadiene (?) water and carbon dioxide, only the first two being in any quantity. Similarly n-hexane gave formaldehyde, ethylene, propylene, $\Delta^{\alpha\beta}$ and $\Delta^{\beta\gamma}$ butylene, two amylenes, three hexylenes, butadiene (?) water and carbon dioxide. The direct oxidation of toluene was studied by Woog[8]. Iron and nickel oxides, copper, manganese dioxide and carbon were investigated as catalysts, and, the proportion of benzaldehyde in relation to the complete combustion recorded. Further work on the contact oxidation of hydrocarbons is put forward by Orloff,[9] who studied Baku petroleum distillates, toluene turpentine, etc. With light petroleum D=0·71, the gaseous products of oxidation included 8–10% carbon dioxide, 6·8–2·4% carbon monoxide, other products being about 10% of closed chain and ethylene hydrocarbons of composition C_6H_{12} and C_5H_{10}, and further aldehydic derivatives of the benzene series with the aldehyde group in the para position. Heavier oils yielded similar products. Toluene, benzene, and turpentine give aldehydic and carboxylic derivatives, and unidentified substances. Natural gas and illuminating gas are quoted as sources of the valuable substance formaldehyde through this process. A typical catalyst in Orloff's researches was freshly reduced copper gauze.

From 1917 onwards a large number of patents appeared mainly from America, covering the formation of valuable intermediates by the direct vapour phase oxidation of aromatic hydrocarbons.

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[5] E.P. 21,941, October 27, 1905.
[6] *Compt. rend.*, 1901, **132**, 1227, 1495.
[7] *Mon.*, 1902, **23**, 773.
[8] *Compt. rend.*, 1907, **145**, 124.
[9] *J. Russ. Phys. Chem. Soc.*, 1908, **40**, 652.

Gibbs[10] covers the oxidation of aromatic hydrocarbons containing one or more side chains or consisting of a number of rings, by treatment of the vapour mixed with an oxygen containing gas (*e.g.* air), at suitable temperatures and in presence of certain catalysts, especially mixtures of the oxides of molybdenum and vanadium. Examples are quoted thus:—toluene vapour mixed with air, passed over vanadium pentoxide between 350 and 450° C., yields benzaldehyde with small amounts of benzoic acid, unconverted toluene and water. Similarly, phthalic acid is obtained from naphthalene, and anthraquinone from anthracene. Fractional condensation is also suggested as an alternative to fractionation of the total product—*e.g.*, with toluene the temperature of the first condenser is adjusted to retain the benzoic acid, the second for benzaldehyde, and the remaining toluene vapour is enriched with air and re-circulated. A similar process was devised by Wohl[11] for the oxidation of naphthalene, anthracene, etc. Benzene as a potential source of maleic acid by partial oxidation and the possible course of the reaction through the intermediate compound quinone is discussed by Weiss and Downs.[12] The pronounced exothermic nature of the reaction is shown by calculation. In the U.S. Patents 1,318,631 and 1,318,632, October 14, 1919, these authors show the conditions and catalysts for the benzene-quinone-maleic acid transformation and vanadium, vanadium pentoxide, molybdenum, tungsten, gold, ruthenium, cobalt, and oxides of copper, manganese, cobalt, lead, chromium, antimony, cadmium and thorium, are quoted. Under fixed conditions a fixed proportion of quinone to maleic acid is formed, which proportion is not altered by introducing some quinone with the benzene. The further patent[13] states the limits of gas mixtures for the latter reaction and also the temperature effect in the formation of maleic acid or diphenyl. Suitable large scale catalyst chambers with alternate catalyst trays and cooling pipes are described by Wade.[14] Selden[15] described a compact form of vanadium pentoxide for a catalyst obtained by sintering the oxide at 685° C. Typical products from pure aromatic hydrocarbons and reaction temperatures are given. Weiss and Downs[16] cover the formation of benzaldehyde and benzoic acid from toluene–air over vanadium pentoxide on pumice at 200–400° C. and also maleic acid if the reaction takes

[10] E.P. 119,517 and 119,518, October 1, 1917.
[11] E.P. 145,071, March 31, 1921, and 156,244, 156,245, April 4, 1922.
[12] *J. Ind. Eng. Chem.*, 1920, **12**, 228.
[13] U.S.P. 1,318,633, October 14, 1919.
[14] E.P. 167,219, April 1, 1920.
[15] E.P. 170,022, April 7, 1920.
[16] U.S.P. 1,374,695, 1,374,720–2, April 12, 1921.

place from 400–550° C. Mercury is used as a heat absorbing agent. For the oxidation of naphthalene, air-hydrocarbon mixtures are passed over aluminium oxide at 350–700° C. Maleic acid is formed also from naphthalene, anthracene, xylene, methyl naphthalene, fluorene and acenaphthene, and its production from toluene is further described by these authors in U.S.P. 1,377,534, May 10, 1921. An interesting discussion of oxide equilibria in catalysis is given by Weiss, Downs and Burns,[17] and it is concluded that an oscillation between higher and lower states of oxidation of the metal supplies the activated oxygen—*e.g.*, between vanadium pentoxide and tetroxide. The preceding patents state facts upon this vapour phase oxidation and indicate only limits of temperature, etc., within which the reaction takes place. An experimental study of the catalytic oxidation of naphthalene using a catalyst of vanadium pentoxide containing small amounts of molybdenum tri-oxide is described by Kusama.[18] More complex catalysts are patented by Meigs and Ellis[19] for the vapour phase oxidation of aromatic hydrocarbons—*e.g.*, iron chromate, chromium vanadate, iron molybdate, cobalt molybdate, cobalt chromate, nickel vanadate, nickel chromate, nickel molybdate, nickel chromate, ceric vanadate, ceric chromate, ceric molybdate, etc., iron vanado chromate, iron ceric chromate, etc. Downs and Stupp[20] cover a technical apparatus for maleic acid production from benzene, also Downs[21] a catalyst chamber with boiling sulphur as a heat absorber. The same author[22] describes the purification of aromatic hydrocarbons by passage over vanadium pentoxide catalyst—*e.g.*, in the case of benzene the impurities thiophene, carbon bisulphide, etc., are oxidised to carbon dioxide, carbon monoxide and sulphur dioxide, while only a little of the benzene is oxidised to quinone and maleic acid, the majority passes over unchanged. Gibbs[23] describes an apparatus for exothermic catalytic reactions, and a mixture of fused sodium and potassium nitrates is used as a heat absorber. Downs[24] reviews the subject of catalytic oxidation in the vapour phase and describes experimental and larger scale apparatus. In the former aluminium is used as a heat absorber, and on graded grains of this metal the catalyst is coated. Early investigators used catalysts that induce complete rather than partial combustion, due to some extent to the heat of reaction. Downs recommends the

[17] *J. Ind. Eng. Chem.*, 1923, **15**, 965.
[18] *J. Chem. Soc. Jap.*, 1923, **44**, 605.
[19] U.S.P. 1,486,781, March 11, 1924.
[20] U.S.P. 1,515,299, November 11, 1924.
[21] U.S.P. 1,589,632.
[22] U.S.P. 1,590,965, July 29, 1926.
[23] U.S.P. 1,599,228, September 7, 1926.
[24] *J.S.C.I.*, 1926, **45**, 188T.

use of an efficient heat absorber for this reaction. In the larger scale plant mercury is used for this purpose. The author reviews also the possible markets for the products such as phthalic anhydride, anthraquinone and maleic acid. Canon and Andrews[25] describe an alloy of cadmium and mercury as an agent for heat absorption in exothermic reactions. Caspari[26] introduces a novel feature in this type of oxidation, insomuch as a colloidal solution of the metals of the fifth group is atomised with air into the heated chamber (200–500° C.). In this way benzaldehyde and benzoic acid may be obtained by spraying a colloidal solution of vanadium oxide in toluene with eight times its vapour volume of air into a chamber at 200–300°. Craven[27] for the production of maleic acid from benzene uses a catalyst of vanadium oxide 70 per cent. and molybdenum oxide 30 per cent., with 0·3 minute contact time. A catalyst carrier is patented by Downs[28] comprising iron particles coated with aluminium. Marks[29] preheats the vapour-air mixtures in catalytic oxidation by mixtures of sodium and potassium nitrates, which also absorb excess heat from the reaction. Downs[30] again reviews the subject of vapour phase oxidation stating some facts and speculations. The formation of anthraquinone, probably by the condensation of some benzaldehyde and benzoic acid, was confirmed by contemporary work of Bowen and Nash,[31] who obtained this condensation not only with toluene but also with toluene-containing petroleum distillates. Gaseous hydrocarbons are also oxidised under these conditions.[32] Methane mixed with 1–3·5 volumes of air is passed at 550° over a catalyst, *e.g.*, molybdic oxide or oxide or compound of a high melting electronegative metal of low atomic volume, *e.g.*, vanadium, manganese, tungsten, uranium, chromium, titanium, and zirconium. Silver oxide can be admixed also. With a contact time of one second formaldehyde and formic acid are produced, the yield being 6·75 per cent. on the methane. The process is applicable to methane homologues, coal gas, and to petroleum still gas, the products in the last case consisting of a mixture of aldehydes and acids.

Further details on the oxidation of heavy mineral hydrocarbons are given by James,[33] in which the heavy oil is mixed with light

[25] U.S.P. 1,614,185, January 11, 1927.
[26] E.P. 263,201, November 11, 1925.
[27] U.S.P. 1,636,857, July 26, 1927.
[28] U.S.P. 1,636,685, July 26, 1927.
[29] E.P. 275,321, May 3, 1926.
[30] *J.S.C.I.*, 1927, **46**, 383T.
[31] *Nature*, 1927, **120**, 621.
[32] James and Byrne, U.S.P. 1,588,836, June 15, 1924.
[33] U.S.P. 1,597,797, August 31, 1926.

petroleum and the mixture vaporised by means of steam. In this way, wax distillate, pressure tar, and heavy lubricating oils have been vaporised and passed through the catalyst screens without clogging them. In the vapour phase the hydrocarbons of high molecular weight are oxidised preferentially. This investigator also suggests the possibility of a combination of vapour phase oxidation and a cracking process.[34]

Allied to this subject is the information contained in the D.S.I.R. Special Report No. 1 (Ledbury and Blair), a report summarising research in the oxidation of hydrocarbons to produce formaldehyde. The oxidation of methane, ethylene and hexane, etc., are described using ozonised oxygen, *e.g.*, ozonised air is passed into boiling n-hexane and products are obtained including: aldehydes (formaldehyde and chiefly acetaldehyde, higher aldehydes up to hexoic), acids (probably all up to hexoic), and in all probability the ester hexyl hexoate.

The résumé given above of previous work indicates that technically the greater part of the research has been devoted to the vapour phase oxidation of pure aromatic hydrocarbons. It was considered of interest to experiment with a toluene-containing petroleum distillate under conditions that caused the conversion of pure toluene into benzaldehyde and benzoic acid. Further the toluol spirit used (containing approximately 59 per cent. toluene) was washed exhaustively with sulphuric acid to remove the aromatic content, and the residual paraffin-naphthene mixture tested under the same oxidation conditions.

It appears that it is not a really economical proposition to obtain a pure toluene distillate from a crude petroleum, but in this connection it is evident that spirits containing approximately 60 per cent. toluene (free from benzene and xylene) were used successfully for nitration purposes during the war.[35]

It is possible that, when such a starting material is used containing aliphatic, naphthenic and aromatic constituents, conditions will be discovered under which certain of these groups can be selectively oxidised. The patents already referred to, in which vapour phase oxidation is used to remove thiophene from benzene, and preferential oxidation of heavier oils, provide evidence in this direction.

Experimental.

For these experiments were used pure toluene (B.D.H.), and a "Special Toluol" Benzine manufactured ex Borneo crude (straight run), supplied through the kindness of Mr. J. Kewley, M.A., F.I.C.

[34] *Chem. and Met. Eng.*, 1922, **26**, 209.
[35] Kewley, *J. Inst Petr. Techn.*, 1921, **7**, 217.

This "toluol benzine" had the following characteristics:—

Specific gravity at 15° C.		0·8115
Colour		Water white
Suspended matter and deposit		Slight water
Odour		Good
Unsaturateds		Negligible
Aromatics		58·9%
Naphthenes (approx.)		30·0%
Paraffins (by diff.)		11·1%
Distillation A.S.T.M. at 755 mm.—		
Initial boiling point		101° C.
Distilling to	102·5	10%
	103	20%
	103·5	30%
	103·5	40%
	103·5	50%
	104	60%
	104·5	70%
	105	80%
	106	90%
	107·5	95%
Final boiling point	120°	98·5%

The apparatus used was based on the experimental design of Downs,[36] except that an electric furnace with a stout iron inner tube was used instead of the metal bath for heating the catalyst. The air was measured in a wet meter and drawn through two scrubbing towers containing respectively caustic potash solution and strong sulphuric acid. It was then passed through a copper worm in the thermostat and bubbled through the liquid under investigation which was also immersed in the thermostat. The vapour-enriched air was then led into the reaction tube, a hard glass tube containing 60–70 c.c. of 5–10 mesh aluminium grains to serve as preheater, and then the catalyst, which consisted in each case of 60 c.c. of similar aluminium grains, but coated with vanadium pentoxide. The exit gases were led through a water-cooled condenser to an ice-cooled flask, and from there through two water washers, and finally two U-tubes filled with activated charcoal.

Oxidation of Toluene.

Toluene, 50 c.c., was run into the "carburettor" which was held at 50° C., and the vapour-enriched air entered the reaction tube at a temperature two degrees lower. Woringer[37] gives the vapour pressure of toluene at 50° C. as 93·53 mm. The catalyst tube was maintained at 400° C. and the gas rate adjusted to approximately 12 litres per hour. The gas-meter readings before and after

[36] *J.S.C.I.*, 1927, **46**, 190T.
[37] *Zeit. Phys. Chem.*, 1900, **34**, 262.

the experiment indicated that 127·5 litres of air were used. At the cool end of the exit tube a mixture of crystals separated, which were divided into 0·30 gm. of crude yellow needles, 4·87 gm. of white flakes and 0·37 gm. of a mixture of both. The reaction water which collected with the oil in the receiver had an acid reaction and decolorised acidic potassium permanganate solution immediately and reduced ammoniacal silver nitrate solution. Weak bromine water was also decolorised. On recrystallisation of the yellow needles from benzene, they showed a m.pt. of 279° C. and a mixed m.pt. with anthraquinone of 278°. From its melting point and properties this substance was identified as anthraquinone. The white flakes, which on recrystallisation from hot water gave a m.pt. of 122° C., were identified as benzoic acid. An oil condensate of 24·7 c.c. and 7·3 c.c. of reaction water were obtained.

The oil condensate was fractionated and 18 c.c. gave 16 c.c. volatile up to 115° C. and a residue of 2 c.c. of crude benzaldehyde (m. pt. of phenylhydrazone 155° C.). The activated charcoal tubes gave 6·5 c.c. of practically unchanged toluene.

Oxidation of "Toluol Benzine."

The catalyst was revivified by heating in a stream of air at 600° C., and an experiment carried out with 50 c.c. toluol benzine under identical conditions as in the case of toluene. Eighty-eight litres of air were used in this oxidation. The products obtained in condensation included 2·255 gm. of crude benzoic acid, and 0·085 gm. of crude anthraquinone, and 0·145 gm. of mixed residue. The 1 c.c. of original benzine remaining in the carburettor was added to the condensed oil making a total of 29·3 c.c. The 9 c.c. colourless oil obtained from the charcoal tubes was not added to the condensed oil before fractionation.

Fractionation of 28 c.c. of the condensed oil gave 26 c.c. distilling up to 117° C. and 2 c.c. crude benzaldehyde. The reaction water (5·7 c.c.) and water from the washers on titration against 0·1 NKOH gave acidity equivalents to 894 m. gm. of KOH and 10·1 m. gm. respectively.

A further experiment with toluol benzine under the same conditions gave substantially similar results. It was noticed also that the reaction water decolorised acidic potassium permanganate, reduced ammoniacal silver nitrate solution and showed a positive iodoform reaction. The washer water showed the same reactions but to a feebler extent. The benzine distilling up to 117° C. from the condensate and the original toluol spirit gave losses of the same order to sulphuric acid of strength approximately 98 per cent.

From this it was inferred that under the experimental conditions employed both aliphatic and aromatic constituents had been attacked.

Another experiment was carried out with the residual spirit after washing the toluol benzine exhaustively with 98 per cent. sulphuric acid and finishing with a rapid wash with 100 per cent. sulphuric acid. This spirit, which should only contain traces of toluene, yielded oxidation products and a larger proportion of reaction water than the previous experiments. The reaction water, besides having a marked acidic action, showed the presence of other oxidation products by its reducing action to acid permanganate and ammoniacal silver nitrate solutions, Schiff's reaction, and the reduction of Fehling's solution on boiling.

N-hexane was found to give oxidation products under similar conditions as were used in the above experiments. Out of the 50 c.c. of this hydrocarbon 45·9 c.c. were recovered as an oil. The reaction water had an acid reaction, gave the iodoform test readily in the cold, using sodium hydroxide, and on warming, using ammonium hydroxide, and exhibited reducing properties indicating the presence of substances containing the aldehyde group.

Summary.

The preliminary experiments described and the survey of the previous work on the vapour-phase oxidation of hydrocarbon are intended for the discussion of the possibility of this process as a contribution to the problem of the utilisation of petroleum as a chemical raw material.

The experiments with a toluene containing distillate show the presence of benzaldehyde, benzoic acid, and anthraquinone among the reaction products.

Some evidence is also put forward upon the oxidation of aliphatic hydrocarbons under the conditions described, which fact would point to the formation of complex mixtures of reaction products when the starting material contains aliphatic and aromatic constituents.

It is suggested that conditions may be controlled so that a selective oxidation can take place when using mixtures and further experiments are being carried out upon this subject.

In conclusion we wish to express our thanks to Mr. J. Kewley for his kindly interest in, and the original suggestion of, this line of research.

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Petrol Engine Lubricants and Lubrication.*

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Lubrication Considered Generally.

The purpose of lubrication is to provide a layer of lubricant between surfaces having motion relative to one another. This prevents excessive wear and substitutes the lower friction between layers of the lubricant for the friction between the dry solids.

The study of lubrication involves an analysis of friction. History says that Leonardo da Vinci was surprised to find that the force required to move a rope coiled up was the same as when the rope was stretched out to its full extent. The area of contact differed in both cases. In 1699, Amontons found that the resistance to relative motion was independent of the area of contact between the moving bodies and varied with the normal pressure, and he concluded that the frictional resistance was due to asperities (projections) on the surfaces. In 1781, Coulomb confirmed Amontons' Law and in 1830 Morin confirmed the conclusions of Coulomb excepting the latter's view of the equality of static and kinetic friction, the former being greater than the latter in Morin's opinion.

There are two types of lubrication, " Boundary " and " Fluid " Lubrication. These are better understood by considering the friction between perfectly clean surfaces and then proceeding to lubricate them. Such surfaces are difficult to produce because they easily acquire greasy contaminating material from the air. When clean surfaces are placed together, the molecules on both surfaces attract each other with a very large force which tends to weld them into a continuous solid. A force of cohesion is operating. Since the molecules in both surface-layers attract each other and a state of equilibrium is soon reached, any force tending to cause slipping will have to move the molecules out of this state of equilibrium and also to rotate them. The friction would be composed of the resistance of the molecules to motion in a straight line and to rotation. Tendency of a force to create relative motion between two perfectly clean surfaces, tends to produce a re-arrangement

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of the molecules in the surface-layers such that the force of attraction is increased, which will increase the force of cohesion and lead to seizure. Motion of a clean watch glass over a clean glass plate marks the latter.

When a lubricant is placed on one of the surfaces, the lubricant saturates the attractive forces of the molecules in the surface layer of the solid, so that when another solid surface is placed on it the capacity for cohesion is considerably lessened. This is the reason why solid surfaces do not weld when they are contaminated in any way. The resistance to motion now depends upon the friction between layers of the contaminant. When the film of lubricant is exceedingly thin, it does not possess the nature of the lubricant as a mass[1]. When an acid such as palmitic acid forms a film on water, the carboxyl groups "dissolve" in the upper layer of the water and the hydrocarbon chain stands up on end perpendicular to the surface. Similar changes take place when a lubricant is applied to a solid surface, viz., there is an arrangement of the molecules of lubricant next to the solid surface.[9] Two solids having a very thin film of lubricant between them, with the molecules in this "upended" condition, are easier to move one over another because of the greater elasticity of the lubricant molecules. The friction is still relatively great because of the rigid way in which the molecules are held by reason of the attraction of the molecules of the solid. When such conditions exist, one has "Boundary Lubrication."

As the layer of lubricant is increased in thickness, the layers of lubricant molecules, outside the range of the attractive forces of the solid molecules, possess the fluidity of the lubricant in a mass of it. When a force is applied so as to promote motion of the surfaces relative to one another, the resistance to motion is that of the internal friction of the lubricant layers situated midway between the two surfaces and is proportional to the viscosity of the oil.[6-7] Viscosity refers to the resistance offered by the lubricant molecules to motion one over the other and is a function of the force with which these molecules attract each other. When such conditions exist, one has "Fluid Lubrication."

In Boundary Conditions of Lubrication, the friction is a function of the chemical constitution of the lubricant and of the nature of the solid surfaces, since these properties affect the attraction between both. A good lubricant for boundary conditions is one which is strongly attracted by the solid—a poor lubricant one which is attracted less strongly.

The differences between both types of lubrication can be illustrated by considering a simple journal bearing. When motionless, the journal and bearing surfaces are in contact, at least they can

only be separated by a film of oil of extremely small thickness. When the journal starts rotating, the friction being of the "solid" type, will be high. As soon as a very thin film of oil gets between the surfaces, the friction will get less, and will be smallest with lubricants composed of fatty acids because of their low co-efficient of friction when in the thin film condition. A hydrocarbon oil containing a small percentage of the higher fatty acids will function almost as well because the fatty acid molecules concentrate on the bearing surfaces. Such oils are advocated for use by Wells and Southcombe,[18] the principle being exploited through the well known "Germ Process" Oils. As the oil film becomes very thick the friction will depend upon the viscosity of the oil at the working temperature.

As the journal revolves, the oil film is established by the dragging action of the surfaces.[14-16] As the speed of rotation increases, the journal is lifted, and floats on a thick oil film. The dragging action also develops a pressure in the film (there is a flow of oil through the ends of the bearing surfaces). When a thick film is established, the fluid film conditions cause the friction to drop to about 100th part of the friction existing during boundary conditions.

The variables entering into the problem of bearing lubrication, have been studied by Barnard and Wilson who indicate that nine variables have to be considered.[2-20] These can be correlated by the equation :—

$$f = \left\{ \frac{zn}{p} \cdot \frac{c}{d} \cdot \frac{l}{d} \cdot S \cdot M \cdot O \right\}$$

f = coefficient of friction.
z = Viscosity of lub. at the working temp.
n = revs. per min.
p = Pressure on bearing.
c = diametrical clearance of bearing.
d = Diameter of bearing.
l = Length of bearing.
S = Surface conditions (kind of metal, smoothness, etc.).
M = Method of oiling and amount of oil.
O = Oiliness factor of the lubricant.

The equation can be simplified to the form

$$f = \frac{k.zn}{p}$$

which points out that an increase in viscosity of the oil, other things being equal, will cause an increase in the friction.

Fig. 1 shows a typical curve obtained by studying the relationship between f, and zn/p. To the right of the minimum friction value, fluid film conditions operate, and to the left those of boundary lubrication. It will be noted that the critical value of zn/p is smaller for a white metal bearing than for one of bronze.

Fig. 2 gives an impression of the disposition of the journal and bearing when running under load, which is exerted by the journal. The point of nearest approach is not vertically below the centre of the journal but to the off side of the bearing. The change in

pressure in the oil film from point to point is important. At PM the pressure is at a maximum and at Pm at a minimum. These points would have to be borne in mind when selecting a spot for oil feed holes and grooves.

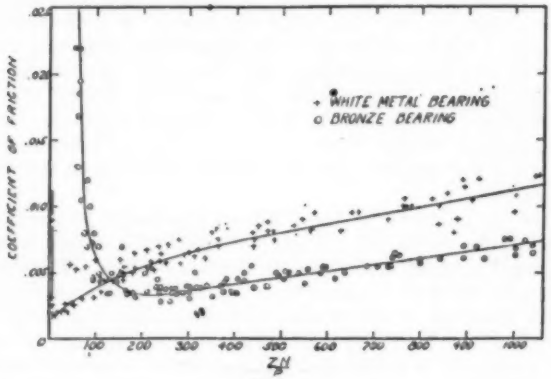

FIG. 1.

The importance of a study of the two types of lubrication is clear when one remembers that the surfaces are subject to boundary lubrication, when they are about to come to rest and when they

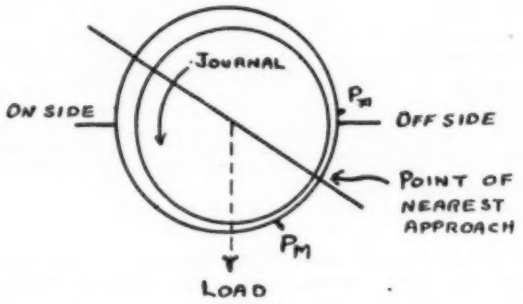

FIG. 2.

just begin to move. The crankshaft main bearings and big end bearings suffer some wear immediately after starting the engine, and when it is coming to rest, but in the case of the piston, the change from one type of lubrication to the other takes place every time it reaches top and bottom dead centres. This does not apply

to sleeve valve engines because the piston and the sleeve always have relative motion even though the piston has to come to a state of rest in a vertical plane.

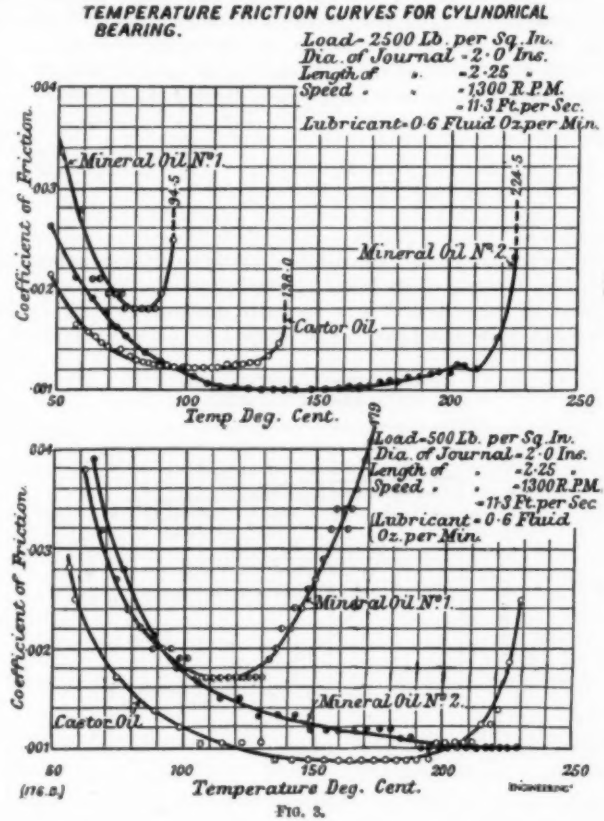

FIG. 3.

Lubrication at elevated temperatures presents difficulties not mentioned hitherto. It is obvious that most oils are very thin at temperatures higher than 100° C. One might be inclined therefore to select a "fatty oil" lubricant or one containing added fatty acids for the purpose in question because of the less danger of abrasion should boundary conditions of lubrication arise. The

confidence might be misplaced because, although these "polar" compounds function favourably in boundary conditions at ordinary or slightly elevated temperatures, it does not necessarily follow that the same success will attend their use at high temperatures. Recent work in academic research indicates this. The most recent contribution to this aspect of lubrication was carried out by Dr. T. E. Stanton at the National Physical Laboratory.[15] The particulars of the oils used are given in the following table :—

TABLE 1.

	Coefficient of friction on the Peeley machine.
Mineral oil No. 1	0·125
„ „ 2	0·126
Castor oil	0·080

These particulars show that castor oil would be superior in boundary conditions because of its lower co-efficient of friction.

These oils were tested in a specially designed bearing. Under high loads, sufficient heat was developed to allow observations to

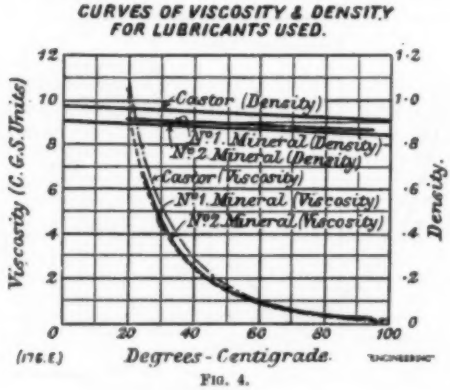

FIG. 4.

be made of the co-efficient of friction over a wide range of temperature. At low loads, the heat was artificially applied. The results are plotted and shown on Figs. 3 and 4, and some of them are tabulated in Table 2.

TABLE 2.

	Min. oil 2.	Castor oil.	Min. oil 1.
Load 2500 lb./sq. in.—			
Minimum friction	0·001	0·0012	0·0018
Seizing temperature	224° C.	138° C.	94·5° C.

This indicates that although one would expect better results from castor oil at higher temperatures, it allowed seizure to take place at a low temperature.

Dr. Stanton draws the following conclusions :—

"(a) That the known merit of castor oil in preventing failure of bearings running hot is not due to inability to seize, this happening in fact at a relatively low temperature. As previously mentioned, it is due to the low value of the coefficient of friction under boundary conditions.

"(b) That it is possible to obtain a straight mineral oil which maintains itself in the film condition up to temperatures far in excess of the limiting temperature for castor oil.

"(c) That mineral oils of the same degree of acidity and unsaturation and for which the values of the viscosity and density are approximately the same, may vary greatly in their capacity to stand up to high temperatures under conditions of film lubrication.

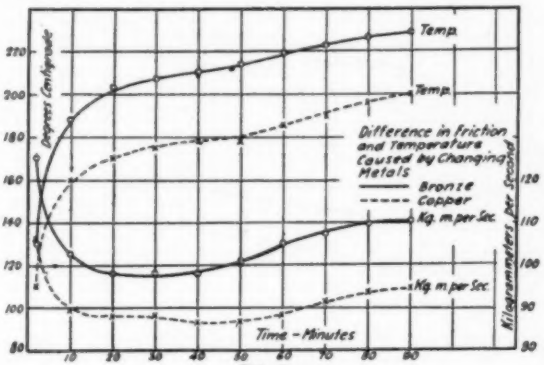

FIG. 5.

"It is clear, therefore, that oils which possess exceptional lubricating properties under boundary conditions are not necessarily the best for purposes of film lubrication and vice versa."

It is also pointed out that the study of both chemical and physical properties of lubricants are important when searching for a lubricant to run at high efficiency and at really high temperatures, but no explanation is given of the remarkably high performance of Mineral Oil No. 2.

It appears possible to offer an explanation which is suggested by the work of E. G. Gilson of the General Electric (America).[5] Gilson measured the friction between a revolving shafting steel

bowl and rings of different metals. A concentric space between both was maintained during the whole experiment so that actual metallic contact was impossible. The standard run lasted 90 mins. The heat generated by friction enabled observations to be made of the power absorbed by friction in the oil over a range of temperature. He also got critical temperatures at which the power absorbed became excessively high. Fig. 5 shows the results for different bearing metals and the same oil. Although in the case of bronze the temperature was higher, i.e., the viscosity of the oil was smaller, the friction was greater than in the case of a copper

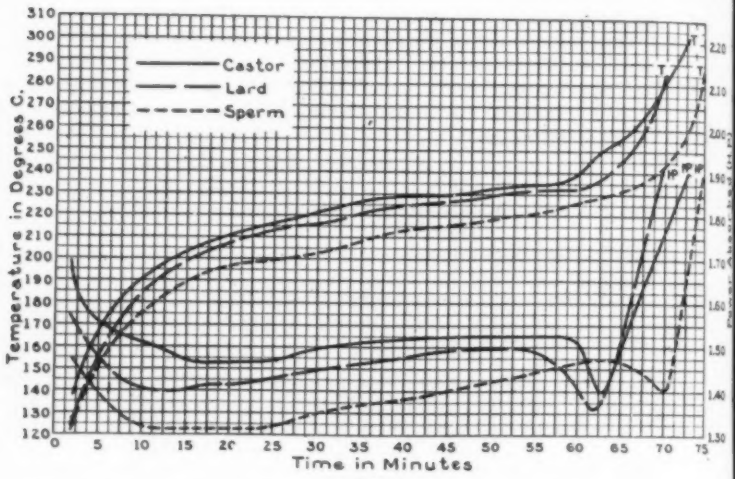

FIG. 6.

Peculiar Dip in Friction and Temperature Curves of Castor, Lard, and Sperm Oils when Run with Copper Rings.

bearing. This relation is contrary to theory. A peculiar factor is exerting a big influence.

Fig. 6 shows results for castor sperm and lard oils. Compared with the previous figure, it will be seen that the temperature rise is steeper for the fatty oils, that they fail to stand up for the standard run of 90 mins., and that they suddenly break down, giving a momentary drop in friction and then they suddenly polymerise to give viscous material which causes an excessive rise in friction. It is interesting to note that sperm oil which contains a large percentage of unsaponifiable matter stands up the longest.

Gilson confirms the fact that polymerisation was the cause of failure of the fatty oils because he describes how the operator had to dig the rubber like contents from the bowl.

With the mineral oils he found a quantity of sludge similar to that formed on oxidation of the mineral oils. In connection with the oxidation of mineral oils, it is interesting to consider his work in another direction. Fig. 7 shows the results obtained for the friction when the experiments were done in an atmosphere of air and hydrogen separately. The steady operating temperature and the friction existing in an atmosphere of hydrogen were higher than in the case of air. When the hydrogen was replaced by

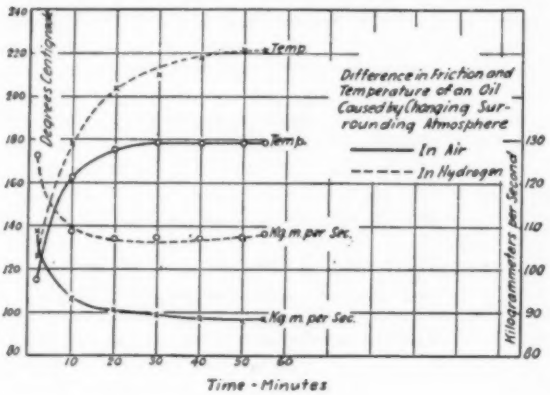

FIG. 7.

oxygen, the friction and temperature quickly dropped to the values obtained when air was used. Fig. 8 shows that the friction was higher in a vacuum than when air surrounded the bearing.

The experiments conclusively show that the presence of oxygen (air) is necessary for a mineral lubricant to give minimun friction and that this low friction is dependent upon, at least, incipient oxidation, i.e., the act of sludge formation. This does not mean to say that all mineral oils would give equal performance because of their tendency to oxidation. Oils which oxidise intensely, will give rise to excessive amounts of sludge-like material and create as much harm as the polymerised products from fatty oils. This work seems to suggest that differences in oxidation tendencies of Stantons Mineral Oils 1 and 2 would explain their different behaviours in the bearing tests referred to.

The question of relative oxidation tendencies of mineral oils affects their use in crankcases, and will be developed in a subsequent section of the paper. The conclusion to be drawn is, that the chemical composition and oxidation tendencies of lubricants derived from petroleum are of importance when the lubricants are to be used at high temperatures.

Petrol Engine Lubrication in Particular.

The most important bearings to be considered are the (*a*) main crankshaft bearings, (*b*) big ends, (*c*) piston and cylinder walls, (*d*) and gudgeon pin. Where splash feed systems are used, all these bearings depend for a supply of lubricant on the rotary motion of the lower parts of the connecting rod, which strike the surface

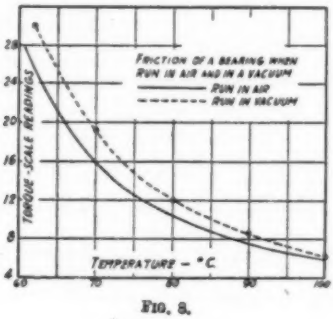

FIG. 8.

of the oil in a constant level trough of oil. The mist so produced has to permeate the whole crankcase in order to get to all the bearings. In forced feed systems, the oil circulating pump drives oil to the main crankshaft bearings, which it lubricates, and part of the oil passes through oil ducts in the crank cheeks to the big end bearings. The oil escaping from these is partly thrown by centrifugal force on to the cylinder walls and on to the lower side of the piston head, thus providing cylinder walls and gudgeon pin with their supply of oil.

The principles governing the selection of an oil for the main bearings and big ends, are those employed in the case of a simple journal bearing, with this difference. In a journal bearing, such as those in electric motors and flywheels, the point of application of the resultant pressure is the same and renewal of the oil film is made only by the dragging action of the surfaces. In a main bearing or crank pin bearing, the point of application of pressure

varies, and this favours easy renewal of oil between the surfaces. The conditions in practice are often more favourable than this, for irregular pressures and change of reaction, causes frequent separation of the journal and bearing, which causes a renewal of oil to be effected. Consequently, these bearings can be expected to sustain several multiples of the maximum pressures permissible in a similar bearing continuously loaded, with the pressure always exerted in one direction. Although it is possible by theoretical means to calculate the viscosity desired for a bearing of known dimensions that has to sustain known pressures, one usually depends upon experience. Consequently, one has only to refer to the charts of engine makers and oils suppliers to find out which grade of oil has the viscosity suitable for a particular unit.

The conditions arising in the bearing surfaces of cylinder and piston are similar to those obtaining for a reciprocating part, such as the crosshead of a steam engine. The conditions are usually satisfied by supplying a liberal amount of oil but, actually, most engine cylinders are over-lubricated. Engines originally possessing this drawback, are now provided with baffles which keep back some of the oil directed on to the walls.

The gudgeon pin bearing has to be classified as an oscillating bearing, and, although the temperature conditions are severe, its lubrication is not so hard to satisfy. R. E. Wilson[19] considers that the oscillating pressure tends to create a very desirable condition. The oil film is not being dragged in at one place and thrown out at the other, but the oil film is kept vibrating. Before it can thin out, the direction of thrust changes, and the oil is pushed back in the opposite direction.

The actual requirements of the different surfaces are difficult to state, but some information on this point is available in the reports of Coleman and Fischer,[4] who studied the "Fresh-Oil System of Lubrication." In the case of an engine of 4½ inches cylinder bore working under full load at 1,500 r.p.m., they found 15 drops of fresh oil per minute gave satisfactory piston and cylinder wall lubrication. On the other hand, only 5 drops of oil per minute were required for each crank pin bearing of a 45-h.p. 4-cylinder truck engine, operating under full load at 1,000 r.p.m.

If the oil is suitable for the demands of the previously mentioned bearings, it should be satisfactory for the lubrication of the other points, viz., the cam-shaft bearings and cams, etc.

If the above mentioned points were the only ones to receive attention when a lubricating oil was being selected, the task would not be as difficult as it actually is. One finds that the oil has to function at much lower temperatures than those of the normal working conditions, i.e., when starting up. This means to say

that the oil is about 7 to 14 times as thick at atmospheric temperature as it is at crankcase temperatures of about 120-140° Fahrenheit, depending on the source of the oil. It is therefore necessary to take into consideration the viscosity of the oil at the average atmospheric temperature for the particular season. The oil should be sufficiently fluid at these low temperatures to enable it quickly and freely to move to the remotest bearings as soon as the engine is started up. The oil should, therefore, be as fluid at these temperatures as possible ; it should also be viscous enough at the normal crankcase temperature and should not be too thin at the cylinder wall temperature. This introduces the question of "viscosity curve." It is well to mention here, that in the case of the piston and cylinder walls, and particularly in those engines having aluminium pistons, the oil is and has to be thicker when starting, so that it can provide an effective seal. The clearances alter with temperature and these are provided for by the varying of viscosity of oils with temperature.

Table 3 shows the possibilities on the oil suppliers' side for a grade of oil which would satisfy The British Engineering Standards Specification for a Light I.C.E. oil.

TABLE 3.

Type of oil.	Paraffin base.		Mixed base.	Naphthene base.		Coal oil from @ N
	Blended	Long residuum.	Mid-Continent.	Russian.	Texas.	low temp. tar
Source.	Pennsylvania.					
Sp. gr. at 60° F. ..	0·8840	0·8795	0·9072	0·9085	0·9290	0·9854
Closed flash point ..	410° F.	410° F.	420° F.	385° F.	365° F.	405° F.
Viscosity at 70° F. ..	885″	850″	1050″	1227″	1435″	1585″
100° F. ..	340″	328″	355″	386″	455″	465″
140° F. ..	127″	125″	127″	127″	127″	120″
200° F. ..	57″	55″	54″	53″	52″	48″
Cold test in ° F. ..	20/25	30/35	30/35	flows 0°	flows 0°	20/25

(The oils are matched on a viscosity of 127 secs. Redwood at 140° F. The L @ N tar oil is included for comparison and not because it is considered to be a desirable crankcase lubricant). The Table is self explanatory. One should mention that the Californian oils which are of a naphthene base are slightly inferior to the Texas oil and would come between it and the L @ N oil.

Other troubles have to be met, such as the formation of deposits in the combustion chamber and crankcase when any lubricant is used. These two aspects should always be considered together, although one finds that experimenters who study one of them, usually ignore the other. It is also true that those who advocate the use of an oil because it gives less cylinder carbon, say nothing about the formation of sludge-like deposits that are formed in the

mass of oil in the crank case, and of the deterioration of the oil itself.

Piston and cylinder head carbon are not in the main what their name implies. Crank case deposits are also heterogeneous masses. The following two analyses illustrate this point.

TABLE 4.

Deposit removed from cylinders of a Ford lorry. Black, soft, lumpy, some hard black particles.	%
"Carbon"	72·1
Oil	18·9
Asphalt	5·7
Mineral matter ..	3·3

Deposit removed from crankcase after big end failure. Black oily paste containing large pieces of "white metal."	%
"Carbon"	36·2
Oil	30·7
Asphalt	3·1
Metal	30·0
(mainly white metal, 11·2% removed by hand)	

Cylinder carbon results mainly from the thermal decomposition of the oil which gets past the piston for a variety of reasons. The oil may be too thin at the cylinder wall temperature, or the rings may have been badly worn. In either case, a large intake manifold depression will cause an influx of oil into the combustion chamber during the intake stroke. This does not apply, of course, to supercharged engines; in these, there is always a positive excess pressure in the combustion chamber during all four cycles. Leakage of oil is inevitable, however, on account of the pumping effect of the rings, and will be excessive when the cylinders are over-lubricated. This would happen if the oil were too thin at the crankcase temperature, since the rate of feed of oil to the cylinders is a function of the viscosity of the oil at this temperature, *i.e.*, in the full splash feed and combined systems of lubrication. Oil that resides in the combustion chamber for any length of time will "crack" and give a "carbon" deposit. Once this forms, it will act as a wick, and assist the creeping of oil from the edges of the piston and the cylinder walls. These deposits are also due in part to oxidation during the intake and compression strokes and when the engine is not working. In these circumstances, the oil is subjected to the action of air at elevated temperatures. This produces oxidised bodies which on decomposition produce more carbon than the unoxidised oil, weight for weight. A fair percentage of the deposits comes from the air, and to a much lesser extent from the fuel. The latter possibility is indicated by the following analysis of material removed from an autovac.

TABLE 5.

Material removed from an Autovac.

	Per cent.
Silica and silicates	6·4
Tin	1·5
Copper	2·8
White lead and lead sulphate	62·0
Iron oxide	10·3
Zinc oxide	0·5
Water and organic matter	16·5

Apart from the water and silica, the other constituents were present in the form of very small particles of paint, which entered the petrol tank when it was filled with petrol from cans. An "Autoklean" strainer fitted between the autovac and the carburettor removed an almost impalpable powder consisting mainly of silica (road dust) which would most probably have passed through the carburettor jet. The importance of strainers in the fuel system and air inlet is great, because they prevent the ingress of material which will cause serious wear to piston rings, etc. Such wear might reflect itself in excessive carbon formation, with an oil which had not previously offended in this direction. Air filters and means of excluding dust from crankcases are also important in the special case of farm tractors, which have to operate in atmospheres heavily laden with grit, when ploughing, harrowing, etc.

A large amount of work has been done in recent years on the relative carbon forming tendencies of oils from different types of crudes.[3-10,11] Table 6 represents the consensus of opinion on this subject, and is taken from the published works of Livingstone, Marley and Gruse.

TABLE 6.

Type of oil tested.	Sp. gr. 60° Baumé °	Viscy. 210° (say) in.	Cold test. ° F.	Per cent. oil in Piston carbon.	Per cent. oil in Head carbon.	Carbon value.	Conradson Carbon value.
Pennsylvanian	30·2	56	—	—	—	7·7	0·51
Mid-Continent	25·2	54	—	—	—	8·8	0·64
Texas Gulf Coast	19·5	49	—	—	—	4·0	0·12
Mid-Continent	24·5	60	5	21·1	14·7	7·3	0·92
Texas Gulf Coast	19·8	60	10	14·1	7·9	4·9	0·22
Mid-Continent	23·8	85	30	17·6	10·0	9·3	1·48
Texas Gulf Coast	18·5	80	45	11·6	6·5	5·3	0·22

(Carbon values column 6 are expressed in grams deposit per litre of oil consumed.)

These results were obtained from carefully controlled test bench work. The same engine was used and the jacket temperature, fuel, level of oil in crank case, temperature of the oil, etc., were kept constant for continuous runs of 15 and 36 hours; in fact, in conditions as different as possible from those of actual practice. The results are of value, however, because they indicate that a relation exists

between them and the Conradson carbon values obtained in the laboratory (by heating a weighed quantity of oil under conditions that are more or less standard). The relation is not a linear one, and is only qualitative. Better comparisons might have been obtained if the Ramsbottom test had been employed because this lends itself to absolute control.

It is generally agreed that it is impossible to get concordant results on carbon formation by road tests, because uncontrollable variables upset all endeavours to keep the tests uniform. (Should the scraper rings, for instance, wear to a greater extent in one engine than in the other, the results would no longer be comparative.) Excessive carbon formation will result if the engine is left running "idle" for a long time. It appears that light load, high piston speeds and small throttle opening all give rise to a large negative pressure above the piston during the intake stroke. The combustion and exhaust pressures are also lower than under heavy load, so that such conditions tend to allow a greater amount of oil to pass the piston. The temperatures in the cylinder also affect the amount of carbon formed. This is generally indicated by laboratory experiments.

TABLE 7.

Effect of temperature on carbon formed at different temperatures.
(Ramsbottom method.)

Temperature.	Per cent. carbon.	
550° C.	0·52	1·36
500° C.	0·85	1·80
450° C.	1·70	2·65

The effect of temperature is also emphasised by Orelup and Lee[12], who describe tests on an engine which was run on light load and partial throttle opening. The cylinder head was removed and showed an accumulation of soft pitchy "carbon." The head was replaced and the engine run under heavy load and full throttle opening. The cylinder heads on removal showed the amount of carbon appreciably diminished.

The question whether paraffin base oils or midcontinent oils give rise to more carbon that the asphalt oils can only be discussed when one has the complete history of the oils. One can get oils from the same crude, having similar viscosity and widely varying carbon forming tendencies, because these depend upon the method and degree of refining to which the oil has been subjected. One cannot refer to oils of different "base" as though they were of constant and uniform composition.

The other difficulties encountered with crankcase oils are connected with the changes in the crankcase. One of these is "dilution," which results in a diminution of the viscosity of the oil. The diluent is derived from the gases leaking past the piston rings

during the compression and combustion strokes. These gases may contain unvaporised fuel, particularly the higher boiling constituents of the petrol, which are absorbed by the oil. In America dilution amounts to 10–15 per cent. in summer, and about 25 per cent. in winter. In this country the dilution is much lower (2–5 per cent.) on account of the better class of petrol used—*i.e.*, it contains less hydrocarbons of low volatility. It can arise also from excessive priming. Dilution reaches a maximum for a particular engine using a uniform quality petrol during any one season of the year. This decrease in viscosity has to be offset by using a new oil with a higher viscosity than is desired, so that when the equilibrium has been reached, the diluted or "equilibrium" oil has the viscosity suited to the engine. One usually finds that excessive dilution leads to trouble very quickly in the form of big end failure, or seizing of equally important bearings. Trouble of this kind would result from changing over from a satisfactory petrol so far as volatility is concerned to a less volatile petrol.

The absorption of diluent by oils of different base has been discussed freely, and if there was any difference it would affect one's selection of the lubricant. The views of Parish[13] prove a useful guide in this connection. He contends that the paraffin base oils will absorb larger amounts of diluent. Against this is offset the recent opinion of Livingstone, Marley and Gruse[10], who maintain that under the conditions used for comparing the carbon-forming tendencies of oils the dilution is the same for all the oils used. It would appear that the disagreement of results lies in the difficulty of getting exactly the same oils for the test work, though the latter view is probably the more correct.

Where dilution occurs water is usually found in the crankcase. The cure of the difficulties of dilution will remove water trouble. The accumulation of water in the crankcase, whatever is its source, will increase corrosion troubles. It will also assist in forming troublesome viscous mixtures with the solid material in the oil, which have been known to clog the oil filters and stop circulation of the oil. The following are analyses of such watery mixtures :—

TABLE 8.

Deposit taken from sump of an A.E.C. bus. Black oily paste containing water.	%	Deposit from sump of Buick engine used in a motor boat.	%
Oil and diluent	43·5	Oil and diluent	57
Water	40·0	Water	36
Asphalt, carbon	13·5	Asphalt, carbon	13·5
Mineral matter (mainly iron, some silica)	3·0	Mineral matter (mainly iron, little copper and tin, some silica)	2·5

The only way to avoid trouble from these troublesome emulsions is to prevent the accumulation of water. Water and diluent can be lessened by operating on leaner mixtures than are most commonly employed; by operating at higher jacket and crankcase temperatures and by attaining normal working temperatures as quickly as possible after starting from cold. All these advantages are claimed for it by the advocates of the "evaporative cooling system."[8]

The other difficulty that arises is that of the formation of solids in the crankcase, and the solution in the oil of products of oxidation of the oil.

A percentage of the solids is formed by decomposition of the oil on the lower side of the piston head, on the inner side of the piston skirt, and on the cylinder walls, to a lesser extent. Analysis of used oil also points to the presence of a considerable amount of asphaltic material produced by oxidation. Conditions for this reaction to go on are most favourable, because the oil is in the form of a mist, at an elevated temperature. The air is also gradually renewed by the "breathing" action common to crankcases, and also one has to include the effect of metals, which, with the possible exception of aluminium, invariably accelerate the oxidation of lubricating oils.

The laboratory study of oxidation leads one to the inevitable conclusion that those oils which contain large amounts of sulphur compounds show the ill-effects of their presence. The writer has not tested any lubricating oil, having high contents of sulphur compounds, which did not give far more oxidised material than a similar oil with less sulphur compounds in it. It is often the case that asphalt formation by oxidation is not a linear function of the amount of sulphur compounds present, because other constituents —*e.g.*, "soft resins," can produced similar results. These bodies can be removed by efficient filtration methods of refining, but sulphur compounds are not always as easy to remove.

An illustration of what befalls the sulphur-bearing compounds of oil on oxidation is afforded by some of the work carried out by C. E. Waters.[17]

TABLE 9.

Oxidation value by Waters Test	4·55
Evaporation loss during oxidation	22·06
Sulphur in original oil	0·46
Sulphur in asphaltic bodies produced by oxidation	2·03
Sulphur in oil freed from asphaltic bodies ..	0·35

These figures indicate that the sulphur accumulates in the asphaltic matter produced during oxidation.

Waters also carried out sulphur tests on the carbon deposits formed when oils of high sulphur content were used in the crankcase. Analyses showed that 12 carbon deposits, freed from oil, had sulphur contents ranging from 0·52 per cent.–3·08 per cent. The portion of the carbon deposit soluble in benzene would contain unchanged oil and asphaltic material. The extracts from 3 deposits give the figures 0·94, 1·33, and 1·52 per cent. for the sulphur content. It is interesting to note that as a lubricating oil is distilled (as it does when giving carbon in engines) the sulphur compounds accumulate in the residue. The carbon deposits represent the last fraction of the destructive distillation of the oil. Consequently, if the oil has oxidised badly in the crankcase, and because these asphaltic bodies are not very volatile, they will concentrate in this residue and cause a larger carbon deposit to be formed than the Conradson Carbon Test of the new oil would indicate.

It would appear, therefore, that the most suitable oil for crankcase work is one having the best viscosity ratio, low carbon-forming tendencies, coupled with comparative freedom from sulphur and a minimum susceptibility to oxidation. All lubricating oils will oxidise, and this is necessary to a certain extent, but excessive oxidation, which is characteristic of certain types of oil, should be avoided.

Satisfying the needs of one wishing to use the ideal oil is about as difficult as supplying the ideal automobile. The latter is considered to be a small light car, odd-looking because it is stream lined, having a high compression engine, a very complex carburettor which only the designer could adjust, an automatic ignition system which will provide the correct spark advance for the particular speed and load, and a small cooling system. It will be made to run only on the lean mixture which gives maximum efficiency, and will be devoid of all powers to accelerate rapidly. The first difficulty that would arise when attempts would be made to sell it would be the extremely high price. Similar difficulties confront the oil refiners. The ideal oil, free from all carbon-forming tendencies, which would have the best viscosity ratio possible, and an extremely low cold test for the type of oil, would be very expensive. A compromise has to be struck between the various requirements of an oil for a particular unit, and this can be done by long-established firms having reputation and standing to maintain.

My thanks are due to Mr. C. Chilvers for carrying out the chemical analyses of the various deposits, and to Mr. L. Gower for compiling Table III, to Mr. J. Wilkie Frye for constant advice and criticism, and to the Anglo-American Oil Co. for permission to publish the analytical data.

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The Relation of the Micelle to the Medium in Asphalt.*

F. J. Nellensteyn, Ph.D.

Asphalt[1] contains three principal groups of components[2]:

1. the medium;
2. a lyophile part: the protective bodies.
3. a lyophobe part: the elementary carbon.

In the system which is formed by these three groups, the disperse phase consists of the two last groups, these being the constituents of the *asphalt micelles*. The stability of the whole system in the first place depends upon the relation between the micelles and the oily medium. Changes in this stability, which are known as flocculative and peptising reactions, are valuable in studying this relation.

The flocculation of the disperse phase of asphalt and other highly protected organosols. When adding benzine of low boiling point to a solution of asphalt in CS_2, C_6H_6, etc., a precipitate is formed. Apparently a new phase appears, and as very little is known about the laws which rule the miscibility relations, there seems to be no reason to expect a simple connection between the flocculating power and other properties of this benzine. Particularly for the organic-chemist the fact that benzine precipitates asphalt solutions must be considered as a purely empiric datum.

As a matter of fact however *no new phase is formed* when flocculating an asphalt solution with benzine, for the asphalt-solution itself is already a two-phase system, as is revealed by the ultramicroscope. This precipitation is simply due to the joining of particles of the disperse phase: it is characterised by changes in the interfacial layers. The precipitation of the asphaltenes must be considered *as a breaking of an "ultraemulsion."* Although in the disperse phase, the asphalt micelle contains elementary carbon, asphalt has a more emulsoid character, the suspension properties being hidden by the protective bodies. A consequence of these considerations is that the stability conditions of macroemulsions may furnish useful data about the factors which rule the peptising and flocculating conditions of asphalt solutions. The extensively investigated oil-in-water emulsions are of great importance in this

* Paper received December 7th, 1927.

[1] The word *asphalt* is used in the American sense as a general name for natural asphalt and residual or blown petroleum products.

[2] See F. J. Nellensteyn, The Constitution of Asphalt, *J. Inst. Petr. Techn.*, 1924, **10**, 311.

respect. For these emulsions H. Limburg gives the following stability-conditions.[3] It depends upon:

(*a*) the electrical charge of the particles;
(*b*) the conductivity of the disperse phase;
(*c*) the interfacial tension;
(*d*) the presence of solid protective layers round the particles of the disperse phase.

It seems probable that for organosols conditions *a*, *b*, and *d* are not of much importance, but that the interfacial tension especially determines the stability of asphalt solutions. An increase of the interfacial tensions decreases the stability. Perhaps the solvatation of the protective bodies, according to the theories of Kruyt[4], also plays a part, but in any case the interfacial tension seems to be the first and most important stability condition. The amount of this tension is unknown, but it may be calculated, with some restrictions given by Antonow,[5] as the difference of the surface tensions of both phases. Thus, by adding a liquid which changes the surface tension of the medium the interfacial tension micelle medium is also changed. If σ_{t1g} and σ_{t2g} are the surface tensions of both phases the interfacial tension $\sigma_{t1t2} = \sigma_{t1g} - \sigma_{t2g}$. Decreasing σ_{t2g} causes an increase of σ_{t1t2}. This occurs when flocculating an asphalt solution; the flocculating reagents have a low surface tension and accordingly the asphalt micelle probably has a high surface tension. Ranging various liquids which are miscible with the medium of asphalt according to their surface tension it is evident that all the low-surface tension liquids are flocculating reagents, whereas those possessing a high surface tension are peptising reagents.

	σ: (dyne/c.M.)	
Ether	17·1	
Benzine 40–60° (from Sumatra)	17·4	Flocculating
n-Hexane	17·4	reagents
Ethylacetate	22·9	
Acetone	23·1	
Butanone	23·6	
Carbon tetrachloride	25·7	Intermediate
Cyclohexane	26·0	zone
Chloroform	26·9	
Benzol	28·8	Peptising
Carbon disulfide	31·0	reagents
Pyridine	35·5	

[3] Onderzoekingen over Emulsies, diss Delft, 1924, 121.
[4] *Z. Physik. Chemie*, 1922, **100**, 250.
[5] *J. chim. phys.*, 1907, 5,372.

$C Cl_4$ and cyclic C_6H_{12} show a surface tension which holds the balance between the values for the flocculating and for the peptising reagents. By this intermediate position these liquids may combine peptising and flocculating properties. They are apt to flocculate especially when a less stable system has been formed. In practice this is caused by faults during the distillation or blowing, or when road asphalt has been overheated during the melting process. A slight decomposition of the micelle when "free carbon" has not yet been formed may be detected by adding $C Cl_4$. *The intermediate position of this liquid as shown by its surface tension explains Richardson's reaction on the so-called carbenes simply as a reaction on a less stable asphalt micelle.*

Cyclohexane has been proposed as a flocculating reagent, by M. Jäkes,[6] who observed a very incomplete precipitation. This could not be confirmed as the cyclohexane used gave no precipitate at all. It is, however, very difficult to get a pure product, and as the surface tension occupies the intermediate zone, a small amount of impurities may greatly influence the flocculating properties.

The high-surface tension liquids are still widely different in their peptising properties. Generally the higher the surface tension the better the solvent power. Asphalts with a high surface tension may contain a coarser and less protected disperse phase than low surface tension asphalts. Mixing asphalts with products of very different surface tension, *e.g.*, coal tar, therefore, sometimes causes troubles. Little is known about the surface tensions of petroleum residues, blown asphalts and coal tars; some determinations were made on this subject and the following numbers found[7]:

Spramex, Mexican residual petroleum asphalt, melt. pt. B. & R. 40°.
$\sigma_{180°}$: 23·5 dyne/c.M.
Shelfalt, Mexican residual petroleum asphalt, melt. pt. B. & R. 65°.
$\sigma_{180°}$: 25·2 dyne/c.M.
Mexican blown asphalt, melt. pt. B. & R. 80°.
$\sigma_{200°}$: 23·1 dyne/c.M.
Mexican blown asphalt, melt. pt. B. & R. 67°.
$\sigma_{200°}$: 23·2 dyne/c.M.
Refined coal tar.
$\sigma_{180°}$: 32·0 dyne/c.M.
Borneo residual petroleum asphalt, melt. pt. R. & B. 63°.
$\sigma_{200°}$: 23·2 dyne/c.M.

The great difference between the values for petroleum and coal-tar products may theoretically explain why many asphalt investiga-

[6] *Chem. Ztg.*, 1923, **47**, 757.

tors, on account of their practical experience, disapprove the mixing of petroleum residues with coal-tar products.

The low surface tension liquids also show great differences in their flocculating properties. The precipitation is never complete; even with a great excess of benzine the finest and most protected micelles are not precipitated. Only by adding liquids which are not totally miscible with the medium or the protective bodies of asphalt may a complete precipitation be achieved. In this case, however, a pure flocculation no longer occurs and the precipitate differs considerably from the flocculated carbon micelle, for it contains a larger portion of oily compounds derived from the medium and the protective bodies. Liquids which are only partly miscible with the medium and the protective bodies of asphalt are *e.g.*, dilute ethanol and acetone, mono-chlor-acetic ester, malonic ester. When precipitating with these liquids we get a "soft asphalt" instead of the "hard asphalt" from the benzine flocculation. In these precipitations the surface tension plays no part.

For the organic chemist hard and soft asphalt, as obtained by these precipitations, are chemically different compounds. From the preceding it follows that the difference between these precipitates is of quite another nature. It is simply the interfacial tension and the miscibility with the oily constituents of asphalt which determines the nature of the precipitated mixture.

In the much discussed question as to which liquid should be used for the determination of asphalt, Holde proposed "normal benzine" for hard, and ether-alcohol for soft, asphalt,[8] Kantorowicz recommended ethylacetate,[9] while F. Schwarz used butanone, and precipitated in the remaining liquid the so-called resins by adding water.[10]

Want of colloid chemical insight involved useless discussions on the supposed superiority of the precipitating methods. It is obvious that none of these methods is an absolute one. They are all conventional comparative methods by which the approximate amount of colloidal carbon and protective bodies can be determined.

When standardising "normal benzine," this liquid was characterised by its gravity and fractional distillation, but it appears that the most important characteristic, *i.e.*, the surface tension, has not been duly taken account of.

Not only asphalt but also other highly protected organosols are flocculated by liquids which increase the interfacial tension between

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[7] *Chem. Weekblad*, 1927, **24**, 57.
[8] *Chem. Ztg.*, 1911, **35**, 369; 1914, **38**, 241, 264, 1,059.
[9] *Chem. Ztg.*, 1913, **37**, 1394, 1438, 1565, 1594.
[10] *Chem. Ztg.*, 1911, **35**, 1417.

micelle and medium. The collodial solutions of NaCl in benzol, for example, are flocculated by ether. Svedberg,[11] who thoroughly investigated these NaCl sols found that when taking ether as a solvent a precipitate was always formed, while benzol gave clear solutions. For these NaCl sols the maximum surface tension which still causes a precipitation is lower than for asphalt. It might be expected that this value should not be equal for all these sols. The flocculated NaCl may be peptised again by benzol, just as is the case with the asphaltenes. In these flocculation reactions the micelle itself is not attacked. If, however, a trace of water is added to the NaCl sol a precipitate is formed, which cannot be dissolved again. In this case the precipitated NaCl contains only a small amount of organic compounds; evidently the micelle has been destroyed. Thus the difference between reversible flocculation and irreversible precipitation by destruction of the micelle may be clearly demonstrated with these NaCl sols.

While flocculating and peptising phenomena are most important in studying the relation between micelle and medium, the irreversible precipitation is prominent with regard to the inner structure of the micelle. It is hoped to deal with these questions in another paper.

Delft, Government Road Materials Chemistry Laboratory.
December 1927.

[11] Die Methoden zur Herstellung kolloider Lösungen anorganischer Stoffe, 1909, 346.

THE INSTITUTION OF PETROLEUM TECHNOLOGISTS.

Rumanian Branch.

FIRST ANNUAL GENERAL MEETING.

THE FIRST ANNUAL GENERAL MEETING of the Rumanian Branch of the Institution of Petroleum Technologists was held at the Industrial International Club, Ploesti, on Friday, October 28, 1927. In the absence of the chairman, Mr. A. Frank Dabell, the chair was taken by Mr. Jacques Iaroslavici.

The annual report and the balance sheet were read and adopted. The Chairman announced that in accordance with the regulations, which provided for the retirement of two members from the committee each year, Mr. F. W. Penny and Mr. J. M. Walsh were nominated and elected for the coming session.

No paper being available, extracts from an article on "Air and Gas Lift," by E. O. Bennett and K. C. Sclater, were read by Mr. Colin M. Leitch, and a general discussion followed.

The meeting concluded with a vote of thanks to Mr. Leitch.

Rumanian Branch.

FIRST ANNUAL REPORT.

The Committee have pleasure in presenting their First Annual Report for the year ended October 28, 1927, together with the accounts to that date.

At the Organisation Meeting the following were elected to constitute the Committee :—

A. FRANK DABELL (*Chairman*).
Captain J. E. TREACY (*Honorary Secretary*).
Captain T. S. MASTERSON, D.S.O.
F. W. PENNY.
JACQUES IAROSLAVICI.
PERCY COWDEROY.

Later, Mr. J. M. Walsh was co-opted to complete the seven members on the committee.

The first financing of the branch was by a donation from the members and non-members interested in petroleum technology.

Later, when the branch was officially organised, the parent Institution made a foundation grant of £5 with a monthly allowance of £2.

The following is a list of members on the branch register on October 28, 1927:—

Honorary Members	1
Members	35
Associate Members	16
Students	3
Associates	1
	56

The average attendance of members and non-members at the meetings for the year was 43.

Eight general meetings of the branch were held during the year under review and the following papers were read and discussed:—

First General Meeting, October 29, 1926: Organisation of the branch and election of the committee. No paper being available the following was read in abstract by Mr. C. M. Leitch: "Oilfield Practice," by A. Beeby Thompson (*Journ. Inst. Petr. Techn.*, 1926, **12**, 351-361).

Second General Meeting, November 26, 1926: "Thermo Dynamics of Some Field and Refinery Problems," by J. T. Hayward.

Third General Meeting, December 30, 1926: "Fuller's Earth and its Uses in the Petroleum Industry," by A. Rauch.

Fourth General Meeting, January 28, 1927: "The Wire Rope," by Percy R. Clark.

Fifth General Meeting, February 25, 1927: "Water Problems in Oil and Gas Fields," by J. M. Walsh.

Sixth General Meeting, March 25, 1927: "Some Notes on Oil Well Casing," by H. L. Little.

Seventh General Meeting, April 29, 1927: "Bean Control of Flowing Wells," by J. F. F. MacQueen.

Eighth General Meeting, May 27, 1927: "Methods of Shutting off Water," by E. J. Dailey.

The committee wish to thank the Industrial International Club for the use of the Club for meetings and also wish to express their appreciation and thanks to Mr. S. Sc. Orescu, Mr. R. W. Davison and Mr. C. M. Leitch for services rendered, and to Mr. Herbert J. Thorne and Mr. Chester M. Guess for acting as Honorary Auditors.

By order of the Committee.

J. E. TREACY,
Honorary Secretary.

REVENUE ACCOUNT FOR THE YEAR ENDED OCTOBER 28, 1927.

Dr.	Lei.		Cr. Lei.
To Printing & Stationery	5,363.00	By Subscriptions ..	4,150.00
To Postage	3,720.50	By Institution of Petroleum Technologists, £25	2,012.500
To Sundry Expenses ..	17,367.00	By Balance	2,175.50
	26,450.50		26,450.50

S. E. & O. Audited and found correct.

HERBERT J. THORNE, M.Inst.P.T.
CHESTER M. GUESS, M.Inst.P.T.

THE INSTITUTION OF PETROLEUM TECHNOLOGISTS

RUMANIAN BRANCH.

FIRST ANNUAL DINNER.

THE First Annual Dinner of the Rumanian Branch of the Institution of Petroleum Technologists was held at the Industrial International Club, Ploesti, on December 2nd, 1927, the chair being occupied by Mr. A. A. Ashworth.

The Chairman (Mr. A. A. Ashworth) said that in March, 1926, some eighteen members of the Institution of Petroleum Technologists gathered together to dine here under the chairmanship of Mr. Frank Dabell, and it was then proposed to form a local branch of the Institution.

As a result of this, early in the summer of last year Dr. Dunstan, a member of the Council of the Institution, was sent to organise this foreign branch and to bring it into active being. This was done, and as a result we see here the first foreign branch of the Institution of Petroleum Technologists.

It is very regrettable that your Chairman, Mr. Frank Dabell, is absent. It is certainly his right to take the chair at this first dinner of the foreign branch which he and his associates helped to institute. In his absence your committee invited me to take his place. At first on receiving this invitation I felt inclined to refuse, partly on account of innate reticence and partly because of doubts as to my standing and capabilities, but I considered it my duty to make the effort as some return for the benefits I had received from the Institution.

This is the first annual dinner of the first foreign branch of the Institution of Petroleum Technologists, and, as such, it must be a notable event in their annals. We have here to-night many guests who are not yet members, and we are honoured by the presence of a representative of the Rumanian Government, and this shows that this branch even in one year has progressed notably.

Now, how is it that a comparatively new institution like that of the Petroleum Technologists has made such wonderful strides in a few years? One is bound to come to the conclusion that the result is due to mutual service. In London one has the advantage of meeting petroleum technologists from all over the world, but

this advantage is practically confined to London, and the real strength of the Institution lies in the service rendered by the publication of its Journal.

We all look to the appearance of the Journal and from it we get fresh knowledge in our own special line and something to help us in our work. I want to ask this branch, if it is to continue as a strong member of the Institution, what it can do to help this work along. The answer should be for all of you to come forward with papers dealing with your own special branch of work. Whenever any of you has anything new, let him bring it forward in a paper and submit it to this branch. All of you have the advantage of being in the forefront of the fight and in touch with new methods in every department, and Rumania is one of the most interesting countries in petroleum science at the present day. If the paper is of general interest, the Institution will publish it in its Journal, and so it will go out to the world and you will be rendering service for service given. I strongly urge each one of you that if you have found anything new you should give a short paper on it to your branch, and in this way you will be doing a great deal to help along the work of producing and refining petroleum.

I would urge you to put everything down on paper—facts and figures; it is little good giving facts and leaving out essential figures. I have seen new refinery equipment described in minute detail but the amount of the throughput for which it was designed was omitted, and such omission leaves me guessing how to gauge the usefulness of the installation.

As this is the first annual dinner of the first foreign branch of the Institution of Petroleum Technologists, I think that you will all join with me when I move that we send a message of greeting to the President and members of the Institution in London, and also that we send a message of greeting to your absent Chairman, Mr. Frank Dabell.

Mr. J. M. Walsh, in proposing the toast "Our Guests," said: The Rumanian branch of the Institution of Petroleum Technologists has been very fortunate ever since its inception in having a large number of friends who have not only taken part in the meetings but have also written and read papers. It goes beyond question that the success of the Rumanian branch has been due largely to the visitors.

The committee is very much gratified with the attendance here to-night. We estimated that 40 would be the maximum and when 73 arrived we were surprised, and thank you.

My particular duty here to-night is to propose the toast of our very distinguished guest who has honoured us with his presence,

but before I get to him I wish to include in the toast the very distinguished friends who have been with us in the year.

It gives me very great pleasure to introduce to you Mr. Gh. Damaschin, who is the Director of Mines, and I am sure you will all give him a very good reception.

Mr. Gheorghi Damaschin, in responding, said: As Director General of Mines I must thank you for your invitation, and transmit to you the greetings of the Minister of Industry, Prof. Mrazec, who was unable to attend personally on account of other obligations.

Outside my official capacity as head of the department which is in direct contact with your activities, I wish to greet you personally and to express my satisfaction at being able to work with you all, who belong to many nationalities and come from various countries in the interest of the Petroleum industry.

I have read the set of rule and regulations of your Institution and I was gratified to recognise that our aims and principles are alike, and that they have for us Rumanian engineers a similar attraction. I also learned that you, as members of the Institution, seek to enlarge your field of knowledge from a technical standpoint.

This purpose goes beyond the horizon of a private association, and I am glad to see the activity of this Institution endeavours to raise the level of its members' knowledge and to find new methods of work to increase the production of oil, and to obtain a good return on the capital invested in the industry.

Our aim being, therefore, a common one, let all countries which possess an oil industry seek to unite their efforts and to collaborate.

I take the liberty of drinking to the health of your President and to the progress of this valued institution.

I conclude with the wish that next year I may speak English and you Rumanian.

Mr. C. M. Leitch, in proposing the toast to the Institution of Petroleum Technologists, said: I believe that the original members who met in 1913 and organised this Institution have inspired their followers with a great enthusiasm. This enthusiasm has generated the local Rumanian branch, and during the past year we had the pleasure of hearing several excellent papers from men who are recognised authorities so far as Rumania is concerned. I should like to make a suggestion that not only should these theses be published in English, but also in Rumanian and in French in some local publication like the "Moniteur du Pétrole," so that they may be read and studied by oil-men who have a knowledge only of French and Rumanian.

I should like to couple with this toast, your late Chairman, Mr. Frank Dabell, our Chairman to-night, and Captain Treacy, and

also Messrs. Russell and Walsh, on account of their efforts in organising this dinner.

Capt. J. E. Treacy said: In reply to the toast to the Institution of Petroleum Technologists, I feel it my duty for the benefit of the many who are interested in Petroleum Technology and the younger members of this Institution to make some remarks regarding the origin of the Institution. To do this I cannot do more than repeat the remarks of our late Honorary Editor, Mr. W. H. Dalton, written in 1915 on the Origin, Progress and Purposes of the Institution, when he said: "The Institution had its inception in the realisation of the need, constantly increasing in urgency, of some means of bringing into contact and co-operation engineers, geologists, chemists and others, engaged in the several branches of their respective professions which are especially connected with petroleum. Even in engineering alone, there is need from time to time of the conjoined abilities and experience of the civil, of the mechanical, and of the mining engineer, so that practically five distinct professions are concerned in the successful conduct of a petroleum undertaking, apart from the commercial ability essential to remunerative expenditure of capital in its development."

Later he says: "Should our anticipation and endeavours duly fructify, the Institution will, as years pass, become a great power of good, not only to its individual members, but to the community at large."

Gentlemen, I believe those anticipations are a reality to-day.

I also refer you to the inaugural meeting of the Institution in 1914, when Sir Boverton Redwood in his presidential address said: "The aims of our Institution are, First, to enable technologists engaged in the petroleum or shale industry to meet and to correspond, and to accumulate trustworthy information regarding the occurrence and production or winning of petroleum and oil shale, the conversion of the raw material into manufactured products, and the character and uses of these products, together with their transport and storage. Second, To promote the better education of persons desirous of becoming professional consulting petroleum technologists, petroleum engineers, geologists or chemists, and to elevate the professional status of those employed in the petroleum industry, by setting up a high standard of scientific and practical proficiency, and by insisting upon the observance of strict rules in regard to professional conduct."

To-day, gentlemen, the institution has the same high ideas and standard as when it was organised.

Of the eighty members who first connected their names with the Institution, probably none realised the future of the organisa-

tion more than Sir Boverton Redwood, and it is doubtful if he conceived the idea that 12 years after its commencement its members would be organising branches in different parts of the world in order to meet and discuss petroleum technology. The Institution was organised in England, but to-day we find it as suggested Mr. Damaschin, more of an international organisation. We have members of all nationalities, and in all parts of the world where oil is to be found. The membership to-day is approximately one thousand.

In England we find two students' sections, one in London and one in Birmingham, where good work is being done.

The first foreign branch to organise was in Rumania, with the able advice of Dr. Dunstan, the honorary editor, who later helped organise the Persian branch. The success of the Rumanian branch we all know, and the work of the Persian branch we can watch with interest from the Journal.

As emphasised by our chairman, the great future of the Institution lies in the Journal, and it is the desire of the honorary editor and us all, that more use be made of the Journal, not only for publication of papers read, with discussions, but for every member to contribute articles on any subject of interest to the industry, especially any advanced ideas, or any new information.

It is our aim to-day, as when the Institution was formed, to further in every way possible the technology of petroleum.

In conclusion, gentlemen, I take this opportunity to thank you for the interest you have shown in our branch.

Mr. J. L. Chaillet said: As a field man, it gives me much pleasure to direct your attention to the activities of the Director General of the Mining Department and to the special studies in production methods that are being conducted there.

As a field man, I should like to emphasise the words of Mr. Damaschin that, in the production department, economy as well as study must be considered, knowledge as well as science. I mean study should not be undertaken merely for the sake of study, but to attain the utmost efficiency for the companies we represent as well as for the country for which we work.

I congratulate Mr. Damaschin on having touched upon that subject, which is bound in the natural course of events to become a major problem for us field men to study. For instance, the economy represented by reducing the cost of casing wells, and preventing gas waste. This latter occurs in all production methods, and one must not decry the real efforts made so far to remedy it, but rather develop a spirit of trying to do better.

I agree with the chairman's remarks that whatever our duties and position may be, we should all contribute in what way we can to the advance of petroleum engineering.

I, therefore, have much pleasure in proposing a toast to the Science of Petroleum Engineering.

Mr. J. Iaroslavici said: As a Rumanian mining engineer, I would like to ask the chairman to express the gratitude of the Rumanian mining engineers to the Institution of Petroleum Technologists for the opportunity it has given them of learning something about the methods which English science has evolved. The methods described in your Journal exemplify modern practice in England and America, and we are glad of the opportunity thus given us of learning about them.

I would like you, Sir, to cordially express our gratitude to the Institution.

Mr. F. W. Penny said: I have one more toast to propose; a short one but an important one, a vote of thanks to our chairman, Mr. A. A. Ashworth, who has consented to take the chair at this dinner. He himself follows the advice he has given us in his opening remarks; he contributes papers and discussions on papers to the Institution. He is also a member of the Standardization Committee of the Institution. We are pleased and honoured to have him here this evening as chairman, and I thank him for the trouble he has taken in presiding.

The vote of thanks was carried by acclamation.

FIELD NOTES, ETC.

Members or others engaged in foreign oilfields and refineries are specially invited to forward notes on any matters of general interest to petroleum technologists. Particulars of any unusual phenomena or occurrences met with by geologists, surveyors, engineers or field operators would be welcomed by the Council with a view to incorporation in the Journal. Photographs or sketches illustrating any descriptions or showing interesting or exceptional occurrences would likewise prove acceptable either for publication or permanent record in the Library of the Institution.

In the Garber Field, Oklahoma, a well is yielding 22,225 barrels of water per day, all of which is run down a dry hole located on the edge of the field. A similar case has been noted at Fort Stockton, Texas, where a well was flowing sulphur water out of the smaller casing and all the water flowed back underground between the outer and inner casing to enter the Trinity sand at the base of the Cretaceous, which was found at a depth of about 200 feet. The sulphur water was coming from the Permian.—SIDNEY POWERS.

In the Little River Field, Seminole County, Oklahoma, a well being drilled in Section 35, T.8.N., R.6.E., is located 660 feet west of another well in Section 36 of the same township. When the former had reached a depth of 2195 feet and the latter 2900 feet, both with rotary tools, the circulation of both wells was lost, and, when one well stopped, mud from the other filled it up. This continued for about two weeks until the drill stems collided and finally a cone from the well in Section 35 was fished out of the well in Section 36.

In the Garber Field, Oklahoma, two wells being drilled 165 feet apart in Section 18, T.22.S., R.3.W., collided when at depths of 3000 feet and 1760 feet respectively, the roller bit of the shallower well cutting a groove in the drill stem of the deeper one.

In the Tonkawa Field, Section 34, T.25N., R.I.W., Kay County, a well drilling at a depth of 1600 feet ran into a well producing at a depth of 2400 feet.—SIDNEY POWERS.

REVIEWS.

LEICHTE KOHLENWASSERSTOFFÖLE. By Dr. Max. Naphtali. M. Krayn, Berlin. Pp. 512+170 illustrations.

This book is the first of a series of six on Hydrocarbon Oils and Fats in course of preparation by Dr. Croner and Dr. Naphtali. The method of dealing with the subject is novel, in that it is based almost entirely on the records of the European and American Patent Offices. The subject is divided and sub-divided into every conceivable ramification, and each subdivision after a few words of introduction consists of a brief account (frequently with diagrams and always with full references) of the principal patents dealing with that particular section of the subject-matter.

Such a mode of treatment gives a book which is naturally incomplete, as only the more important basic patents can be included. On the other hand the author has exercised great discrimination, so that the great masses of patents which consist of trivial variants of known and accepted processes or of claims which any chemist at once realises are fundamentally unsound have been discarded, while the name index shows that the great majority of reputable patented processes have been included.

The book is divided into two main sections dealing respectively with the light hydrocarbons obtainable from petroleum and from coal.

The first section is subdivided into eight sub-sections, the first six of which deal with cracking under various conditions, the seventh with the production of hydrocarbons from natural gas, shale and oil sands, and the eighth with anti-knock motor fuels.

The second section is concerned almost entirely with the Bergius process, the Fischer and allied processes and the various methods and plants for the low temperature carbonisation of coal.

A third and very small section deals with the refining of cracked products and the conversion of the gaseous olefines into alcohols.

The index, which is one of the most important parts of such a reference book, has been well compiled, and the volume is one which will prove of great value to many petroleum technologists, particularly those interested in cracking and in the conversion of coal into liquid fuels. F. B. THOLE.

THE THEORY OF EMULSIONS AND THEIR TECHNICAL TREATMENT. By William Clayton, D.Sc., F.I.C. (London, 1928. J. & A. Churchill. Pp. xi+283.)

The first edition of this work appeared in 1923 under the title "The Theory of Emulsions and Emulsification," and filled a very distinct gap in the literature of disperse systems. In the intervening period a considerable amount of work has been done, especially by American investigators, the inclusion of which as well as a much extended treatment of the technical aspects of the subject has enlarged the volume by over hundred pages.

Among the new investigations described briefly, but adequately, by the author, those of Seifriz are of particular—if painful—interest, as they show an unexpected influence of the density of mineral oils, otherwise quite similar in character, on the type of emulsion formed with the same emulsifying agent. As Dr. Clayton observes, these results "will have to be taken into

account in any proposed general theory of emulsions," but the task of doing this might give pause to the most gallant speculator.

Other new chapters describe the more recent methods of determining surface and interfacial tension, and the very important developments in finding the size-frequency distribution. Lecomte de Nouy's apparatus should supersede the older contrivances for measuring inter-facial tensions: it is applicable where the drop number method—apart from its other known defects—fails, viz. when the two phases have very nearly the same density. The size-frequency curve is really the only method of comparing emulsions or the efficacy of different emulsifying agents, and it is difficult to overrate its importance; the next step will have to be a method for emulsions, the phases of which have nearly the same density, of which there are some quite important examples in industry.

In an appendix, Prof. W. Ramsden sketches a theory of emulsions stabilised by solid particles; unfortunately the theory of capillarity on which it is based has not yet been published in extenso. A brief outline of it is, therefore, given, which appears to contain some points on which opinions are likely to differ. The only safe course is to defer discussion—which is in any case beyond the scope of this review—until a fuller statement of the whole theory is available.

A bibliography, filling 24 pages, in which the reviewer has not been able to detect omissions of any importance, completes the book. The volume is excellently printed, illustrated and bound, and will be indispensable to those who have to deal with the interesting and troublesome systems which form its subject.

E. HATSCHEK.

LUBRICATING GREASES. By E. N. Klemgard, B.Sc. Publishers: The Chemical Catalog Company, Inc., 419, Fourth Avenue, at 29th Street, New York, U.S.A.

This book represents apparently the first attempt so far made to devote an entire work to the subject of Lubricating Greases. The rapid developments in the manufacture of these products in recent years, and the increase of popularity in their use, certainly make the industry of sufficient importance to merit its own text book.

In the chapters dealing with cup greases in which lime soaps are the base, after a discussion of the theory of their composition, full instructions are given of the plant required for manufacture, with many formulæ and detailed instructions for their actual manufacture both in open pans and by the pressure system, which system incidentally is considered to be the system of the future. The wide application of these greases is shown in a remarkable table which is published, comprising a list showing over 250 different purposes for which they are used in industry, together with the approximate grades of grease most suitable for the purpose, their penetration tests, melting points, and the viscosity of the oils to be used in compounding. The proposed A.S.T.M. methods of analysis for these greases are also included, and will be of assistance to the analyst who has usually to devise his own methods suitable for dealing with the product under investigation.

The soda base greases—*i.e.*, gear grease and solidified oils, etc.—receive similar full treatment, and notes are also included of the patent literature dealing with their manufacture. A good word is put in for the mixed soda lime base greases, in which the ratio of lime to soda is usually 1 : 3, and which have the smooth buttery texture of the lime greases but are characterised by the remarkable stability and high melting point.

In the chapter on the cheaper grades of semi-solid grease—viz., the lime soap rosin sett greases—a valuable research is recorded in detail, which was undertaken to investigate the cause of the well-known difficulties in uniform production of these greases owing to the variation in the quality of rosin oil obtainable. The results show pretty clearly that the amount of acidity of the rosin oil is not necessarily the most important factor, but that it is the nature of the acids present, which are decomposition products of the ordinary rosin acid, which varies most and gives the varying results in manufacture.

There are further chapters dealing with special greases and compounded oils in which lead, zinc and aluminium are used as the base, and some mention is also made of cutting oils and soluble oils, and at the end of the book there is a chapter indicating the trend of future research on the subject.

In conclusion, it must be said that if experience proves that the formulæ and instructions are good ones, the book must in all respects be considered an admirable effort. At the same time, the fact remains that grease making is so much an art that it is doubtful if it is possible to teach it in any book. Nevertheless, there can scarcely be anyone in the trade concerned who will not find something suggestive and helpful from the study of its contents.

A. RAYNER.

BOOKS RECEIVED.

LA LÉGISLATION FRANÇAIS DU PÉTROLE. A. de Boulard. Paris, La Revue Pétrolifère, 1927. 30 francs.

This work contains the French laws regarding petroleum and its products. The first chapter deals with the various mining laws from 1810 to date, and the following five chapters give the regulations and decrees covering imports, specifications and testing, manufacture, warehousing and sale, Customs and fiscal.

LANCASHIRE COALFIELD. THE WIGAN FOUR-FEET SEAM. Dept. of Scientific and Industrial Research. London, H.M. Stat. Office, 1927. Pp. 60. 3s. net.

This is the tenth of the series on the physical and chemical survey of the national coal resources, and describes in detail the results of laboratory experiments carried out on the Wigan Four-feet Seam of the Lancashire Coalfield.

THE REACTIVITY OF COKE. I.—STANDARDISED METHOD FOR THE DETERMINATION OF COMPARATIVE VALUES. J. H. Jones, J. G. King and F. S. Sinnatt. London, H.M. Stat. Office, 1927. Pp. 32. 1s. net.

An investigation is in progress at the Fuel Research Station into the reactivity of coke, and this paper describes an apparatus designed to measure the amount of carbon dioxide reduced when the gas is passed over heated coke under standardised conditions. Some results are given, the numerical values for the "reactivity" of various cokes being comparable, but only in reference to the precise conditions specified.

THE "UNSATURATED HYDROCARBONS" IN THE GASES FROM THE CARBONISATION OF COAL. A. B. Manning, J. G. King and F. S. Sinnatt. London: H.M. Stationery Office, 1928. Pp. 19. 6d. net.

This work describes a method devised for the determination of the compounds in the gases from the carbonisation of coal, and gives results obtained in both high- and low-temperature gases.

ERRATA.

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